An air pollution modeling system for Switzerland using WRF-Chem
-
development, simulation, evaluation

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Abstract

Air pollution has become one of the world's most concerning environmental problems. The Swiss cohort study on air pollution and lung diseases in adults (SAPALDIA) examines long-term health effects of air pollution and other environmental and meteorological conditions in Switzerland. Health surveys were conducted in the years 1991, 2002 and 2010. This work contributes to SAPALDIA by developing, simulating and evaluating a new approach for population based air pollution exposure assessment. The approach is based on a chemical transport model (CTM) with an on-line implementation of the chemical reactions to a numerical weather forecast (NWP) model. The Weather Research and Forecasting (WRF) model with its chemistry extension (WRF-Chem) has been applied to the whole of Switzerland with a horizontal resolution of 2 km. This high resolution domain is nested into a coarser European domain to have the meteorological as well as the chemical initial and boundary conditions. For the initialization of the coarser European domain itself, different chemical and meteorological boundary and initial conditions have been evaluated. Anthropogenic emissions were built on the basis of Federal emissions for PM_{10}, $PM_{2.5}$ and NO_x.

The evaluation of spatial and temporal distribution of O_3, NO_2, NO, PM_{10} and $PM_{2.5}$ revealed systematic bias. Post-processing routines with multi-linear regressions eliminated such biases and finally achieved satisfactory results. Yearly averages of air pollutants were simulated with Pearson correlation coefficients up to 0.8. Seasonal trends and spatial distribution are captured correctly by the modeling system. For example, a strong PM_{10} advection from northern Italy could be shown. Outliers of measurement stations compared to WRF-Chem are explainable by the geographical parameters of the station. Compared to a dedicated dispersion model (PolluMap), WRF-Chem achieved roughly the same statistical values for PM_{10} and slightly lower ones for NO_2. However, WRF-Chem is able to produce temporally refined output for more pollutants. Modeled diurnal cycles showed smaller amplitudes as measurements. The necessary daily peak values for violations of air quality standards as set by the Swiss government and the European commission can not be reproduced due to the spatial resolution. Overall, WRF-Chem produced useful yearly averages of air pollutants for exposure assessment.

Contents

A1: Ritter et al. (2012), *Meteorol. Atmos. Phys.* [**in press**]

A2: Ritter et al. (2013), *Atmos. Res.* [**in review**]

A2: Ritter et al. (2013), *Atmos. Res.* **[in review]**

List of Figures

A1: Ritter et al. (2012), *Meteorol. Atmos. Phys.* **[in press]**

A1: Ritter et al. (2012), *Meteorol. Atmos. Phys.* [**in press**]
A2: Ritter et al. (2013), *Atmos. Res.* [**in review**]

List of Tables

A1: Ritter et al. (2012), *Meteorol. Atmos. Phys.* [**in press**]
A2: Ritter et al. (2013), *Atmos. Res.* [**in review**]

Acronyms

ARW	Advanced Research WRF Solver
BSC	Barcelona Supercomputing Center
CAMx	Comprehensive Air Quality Model with Extension
CBM4	Carbon Bond Mechanism Version 4
CBM-Z	Carbon Bond Mechanism Version Z
Cercl'Air	Swiss Society of the Responsible for Air Pollution Control
CF	Conversion Factor
CFD	Computational Fluid Dynamics
CLRTAP-UNECE	Convention on Long-range Transboundary Air Pollution - United Nations Economic Commission for Europe
CMAQ	Community Multiscale Air Quality Model
COSMO-ART	Consortium for Small-scale Modeling - Aerosol and Reactive Trace Gases
COST	European Cooperation in Science and Technology
CPU	Central Processing Unit
CWF	Chemical Weather Forecasting
EMEP	European Monitoring and Evaluation Program
FOEN	Federal Office for the Environment
GEM-AQ	Global Environmental Multiscale Model - Air Quality
GENEMIS	Generation and Evaluation of Emission Data
GFS	Global Forecast System
Enviro-HIRLAM	Environmental High Resolution Limited Area Model
IC/BC	Initial Conditions / Boundary Conditions
LMDZ-INCA	Laboratoire de Météorologie Dynamique Model Version Z - Interaction with Chemistry and Aerosol Model
LUR	Landuse Regression
MADE	Modal Aerosol Dynamics Model for Europe
MAGE	Mean Absolute Gross Error
Map3D	Mesoscale Air Pollution 3D Modelling
MB	Mean Bias
MCCM	Mesoscale Climate Chemistry Model
MCR	Meteorology, Climatology and Remote Sensing - University of Basel
MEGAN	Model of Emissions of Gases and Aerosols from Nature
Meso-NH-C	Mesoscale Non-Hydrostatic Model with Chemistry
MetPhoMod	Meteorology and Atmospheric Photochemistry Mesoscale Model
MOS	Model Output Statistics
MOSAIC	Model for Simulating Aerosol Interactions and Chemistry
MPI	Message Passing Interface
NABEL	National Air Pollution Monitoring Network
NALROM	NOAA Aeronomy Lab Regional Oxidant Model
NCAR	National Center for Atmospheric Research
NCEP	National Centers for Environmental Prediction
NetCDF	Network Common Data Form
NMM	Nonhydrostatic Mesoscale Model
NOAA	National Oceanic and Atmospheric Administration
NWP	Numerical Weather Prediction
PSI	Paul Scherrer Institute
R	Pearson Correlation Coefficient

RMSE	Root Mean Square Error
RRTM	Rapid Radiative Transfer Model
SAPALDIA	Swiss Cohort Study on Air Pollution and Lung Diseases in Adults
SNAP	Selected Nomenclature for Reporting of Air Pollutants
SOA	Secondary Organic Aerosol
SwissPRTR	Swiss Pollutant Release and Transfer Register
SwissTPH	Swiss Tropical and Public Health Institute
USGS	United States Geological Survey
WHO	World Health Organization
WPS	WRF Preprocessing System
WRF	Weather Research and Forecasting
WRF-Chem	Weather Research and Forecasting Model with Chemistry Extension

Molecular formulas

CO	Carbon Monoxide
HCHO	Formaldehyde
HNO_3	Nitric Acid
HONO	Nitrous Acid
NH_3	Ammonia
NMVOC	Non-methane Volatile Organic Compounds
NO	Nitric Oxide / Nitrogen Monoxide
NO_2	Nitrogen Dioxide
NO_3	Nitrate Radical
NO_x	Nitrogen Oxide
O_3	Ozone
PM_{10}	Particulate Matter $< 10\ \mu$m
$PM_{2.5}$	Particulate Matter $< 2.5\ \mu$m
PM_{coarse}	Particulate Matter $< 10\ \mu$m and $> 2.5\ \mu$m
SO_2	Sulfur Dioxide
SO_x	Sulfur Oxide
TSP	Total Suspended Particulates

1. Introduction

1.1. Preface

Air pollution has become one of the world's most concerning environmental problems. With industrialization, anthropogenic emissions increased and consequently air quality decreased rapidly. In the last decades many health studies were conducted with alarming results. According to the World Health Organization (WHO) there is a significant risk factor for multiple health conditions caused by air pollution (WHO, 2006). Long-term exposure to high air pollution concentrations is especially harmful. Air pollution can be linked to reduced lung capacity and respiratory infections (Ackermann-Liebrich et al., 1997; Gauderman et al., 2004), elevated risk of heart disease and mortality (Dockery et al., 1993; Filleul et al., 2005; Jerret et al., 2005; Krewski et al., 2005; Künzli et al., 2000; Pope et al., 2002; Gehring et al., 2006) and lung cancer (Nyberg et al., 2000; Vineis et al., 2006). The impact of these health effects are strongly dependent on the individual health status and genetics as well as the duration, degree and type of pollution a person is exposed to. The most studied and most common sources of pollutants are particulate matter, ozone, nitrogen oxides and sulfur dioxide. Over 850,000 deaths per year worldwide are caused by outdoor air pollution (WHO, 2007). Therefore, legal regulations were introduced to set maximum atmospheric concentrations for specific pollutants. Air quality guidelines are described in WHO (2006) and European legislation are listed in EU (2008). Switzerland also has a legal ordinance for air quality standards.

1.2. Motivation and objectives

One of the above mentioned studies on air pollution and health effects is the Swiss cohort study on air pollution and lung diseases in adults (SAPALDIA). This epidemiological cohort study examined long-term health effects of air pollution and other environmental and meteorological conditions in Switzerland (Ackermann-Liebrich et al., 1997, 2005; Bayer-Oglesby et al., 2006; Liu et al., 2007, 2012). It was initiated in 1991 with health examination follow-ups in 2002 and 2010. Over 9,500 subjects participated in 1991. The same participants were asked to continue their health updates in the follow-up years. In 2010 still over 6,000 subjects have taken part in SAPALDIA. The study was initiated in eight Swiss study areas (Geneva, Basel, Lugano, Aarau, Wald, Payerne, Davos and Montana). As all subjects lived in one of these areas, the first approach for exposure estimates based on measurements. However, over the last two decades many subjects moved house and dispersed all over Switzerland. As it is not possible to make over 6,000 individual measurements, new exposure estimation methods had to be applied. Liu et al. (2007) describes the different approaches so far used for SAPALDIA. The first approaches based on central-site measurements to statistical models and finally dedicated dispersion models. However, all of these kind of models have their strengths

and weaknesses. The most important limitation of all these approaches is the lack of the correct representation of meteorological parameters. The modeling of within-city difference with very high horizontal resolution for the eight areas have been done with hybrid landuse regression models (LUR) including dispersion modeling results as well as geographical, temporal and meteorological parameters (Liu et al., 2012). However, these hybrid LUR models can not be applied over Switzerland as a whole. Therefore there was a need for a sophisticated chemical transport model (CTM) including the physical simulation of meteorological parameters as well as parameterized chemical reactions of the pollutants. This study is the result of such an approach. It has been made possible through a co-operation of the Swiss Tropical and Public Health Institute (SwissTPH, former Institute of Social and Preventive Medicine, University of Basel) and the research group of meteorology, climatology and remote sensing (MCR) from the University of Basel. The overall aim of this study and this thesis is to develop, simulate and evaluate a new, sophisticated approach of exposure estimations for all SAPALDIA participants for the years 1991, 2002 and 2010; this means the application of a CTM for the whole of Switzerland.

1.3. State of current research

Air pollution exposure estimations were often based on measurements in the past. Either central-site measurements or statistical models using measurements were the most common approach. The introduction of personal computers allowed the development of more sophisticated, dedicated dispersion models. These models have no or only few chemical reactions incorporated and are only applicable to very few pollutants. During the last decades the increased availability of computational resources increased drastically. The research field of numerical weather prediction (NWP) benefited from these computational resources and achieves remarkable result simulating meteorological parameters (Jacobson, 2005). As a consequence, there are reliable tools for simulating atmospheric motions and parameters, which are important for air pollution modeling. The implementation of parameterized chemical reactions into a model and the use of modeled meteorological parameters from NWP's as input datasets are the consequence. Such models are often called chemical transport models (CTM) as well as air quality models or air chemistry models (Kukkonen et al., 2012, 2009; Seinfeld and Pandis, 2006). CTM's disperse pollutants with wind vectors and are actually calculating chemical reactions, which separates them from dedicated dispersion models. Depending on various factors, CTM's can be applied for regional to global scales. If the study focus is more on the local scale, LUR's or Computational Fluid Dynamics (CFD) models are more adapted. However, for modeling entire Switzerland, only a CTM is suitable.

1.3.1. Chemical transport models

Chemical transport models can be divided into two different groups depending on the interaction of the NWP and the CTM (Kukkonen et al., 2009; Baklanov et al., 2007; Grell et al., 2005). Off-line modeling systems use two completely different models. On the one hand a NWP and on the other hand a CTM that uses the output of the NWP model. Because of this, off-line models are also called one-way interaction models. On the other hand on-line modeling systems consist of only one overall model. This means that on-line models simulate meteorological parameters as well as the chemical parameters. A schematic diagram of the two approaches can be seen in Fig. 1.

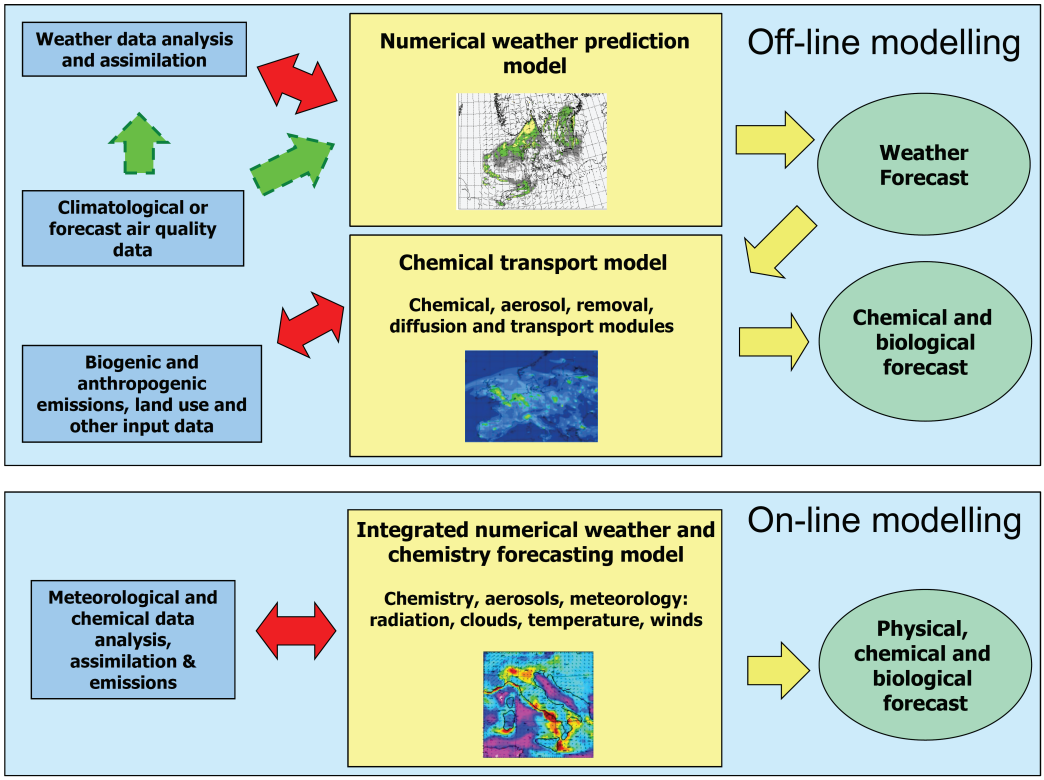

Figure 1: Schematic diagram of the off-line and on-line coupled NWP and CTM modeling approaches. From Kukkonen et al. (2012).

Both approaches have their advantages. Off-line models can have different mesh grids than the NWP models. As most of the studies only have an interest in near ground air pollution concentrations, the off-line CTM can have less vertical layers than the NWP and therefore save a significant amount of computing resources and time. However, they need a special interface module to transform the meteorological output from NWP's into the needed format. Every additional transformation increases systematical errors.

As the off-line CTM cannot interact with the NWP, the model can only interact in one way. On-line models on the other side can interact in both ways and are therefore called two-way interactive models. The parameterized chemical reactions have to be calculated on every grid point of on-line models which leads to high demands on computing resources. However, no temporal or spatial conversion steps are needed and chemical components are fully consistent with meteorological components.

Currently, off-line models are common than on-line models. However, the development of on-line models is more pursued at the moment (Grell and Baklanov, 2011*a*). Tab. 1 shows the most applied off-line models in Europe as well as some available on-line CTM's. The first three models are used by far the most by the European air quality modeling community. The Comprehensive Air Quality Model with Extension (CAMx) has been used amongst others by Couach et al. (2004), Astitha et al. (2008) and Hirtl and Krüeger (2010) for studies in Switzerland, Greece and Austria. The Community Multiscale Air Quality model (CMAQ) is an active open-source project of the U.S. EPA Atmospheric Science Modeling Division. It has been applied to Europe or a subset of it by van Loon et al. (2004), Baldasano et al. (2008*b*), San José et al. (2008) and Matthias (2008). The CHIMERE model is developed in France and therefore widely used for European domains (Schmidt et al., 2001; Bessagnet et al., 2004; van Loon et al., 2004, 2007; Vautard et al., 2005; Baldasano et al., 2008*b*; Szopa et al., 2009; Pay et al., 2010). A lot of other off-line CTM's exist and have been used (van Loon et al., 2004, 2007; Pay et al., 2010; Kukkonen et al., 2012). From the beginning this study focused on applying an on-line model rather than an off-line one. On one hand because it is a novelty to apply a on-line CTM for health exposure assessment and on the other hand because future developments are tending to on-line models.

Table 1: Commonly used off- and on-line chemical transport models.

CTM	Coupling	Homepage
CAMx	off-line	`http://www.camx.com`
CMAQ	off-line	`http://www.cmaq-model.org`
CHIMERE	off-line	`http://www.lmd.polytechnique.fr/chimere/`
Unified EMEP	off-line	`http://www.emep.int/OpenSource/`
WRF-Chem	on-line	`http://ruc.noaa.gov/wrf/WG11/`
Enviro-HIRLAM	on-line	`http://www.hirlam.org/`
COSMO-ART	on-line	`http://www.imk-tro.kit.edu/3509.php`
GEM-AQ	on-line	`http://collaboration.cmc.ec.gc.ca/science/rpn/gef_html_public/`
Meso-NH-C	on-line	`http://mesonh.aero.obs-mip.fr/mesonh/`
MCCM	on-line	`http://imk-ifu.fzk.de/485.php`

From the on-line models, the Weather Research and Forecasting (WRF) model with its chemistry extension (WRF-Chem) is the most frequently used globally. It is freely available and under strong development by several national agencies in the U.S. and worldwide (Grell et al., 2005; Fast et al., 2006; Skamarock et al., 2008*a*). It has been applied for European domains or sub-domains by San José et al. (2008), Schürmann et al. (2009) and Tuccella et al. (2012). The Environmental High Resolution Limited Area Model (Enviro-HIRLAM) is also commonly used, especially for countries who contribute to the development (Denmark, Estonia, Finland, Iceland, Ireland, Netherlands, Norway, Spain, Sweden, Lithuania and France). Korsholm et al. (2008) and Baklanov et al. (2008); Baklanov and Nuterman (2009) describe the model in detail. Germany, Switzerland, Italy, Greece, Poland, Romania and Russia were mainly the driving force for the development of the Consortium for Small-scale Modelling - Aerosol and Reactive Trace gases (COSMO-ART) model (Vogel et al., 2009; Knote et al., 2011). Other on-line models are the Global Environmental Multiscale model - Air quality (GEM-AQ) developed in Canada (Kaminski et al., 2008), the Mesoscale Non-Hydrostatic model with Chemistry (Meso-NH-C) developed in France (Tulet et al., 1999, 2003, 2005) and the Mesoscale Climate Chemistry Model (MCCM) which is kind of predecessor of WRF-Chem and was developed in Germany (Grell et al., 2000). A good overview of different on-line models can be found in Baklanov et al. (2011).

As we see, several countries developed different on-line models in parallel. However, a COST Action (European Cooperation in Science and Technology) has been launched for focusing on a new generation of on-line CTM models. The COST Action (ES1004) is an European framework for on-line integrated air quality and meteorology modeling. It is linked to the COST Action ES0602 (Kukkonen et al., 2009). Both COST Actions started after the beginning of this thesis.

For this study, the best option as CTM was the WRF-Chem model. As previously stated, this study aimed to apply an on-line model from the beginning. The decision for WRF-Chem was not only taken because the model is freely available but mainly due to the existing expertise of WRF at MCR and the modular approach of WRF-Chem. The model and its advantages are elaborated in Chap. 2. When this study was initialized in 2007 it was one of the first times that WRF-Chem was applied to Europe (and Switzerland) for a full year evaluation. However, in the meantime other studies were conducted over Europe using WRF-Chem (San José et al., 2008; Schürmann et al., 2009; Tuccella et al., 2012).

1.3.2. Air pollution modeling systems

An air pollution modeling system is actually the application of a CTM. Kukkonen et al. (2012) defined such systems as chemical weather forecasting (CWF) models as these systems can be used for operational forecasting purposes. Every CTM needs initial and boundary conditions (IC/BC's) as well as anthropogenic and biogenic emissions of

pollutants. The preparation of all needed datasets, the application of the CTM and the eventual post-processing are summarized as an air pollution modeling system. Such systems often have several nested domains to achieve high resolution modeling. The coarsest domain uses data from global chemical and meteorological models as input. The most critical point of such air pollution modeling systems are the anthropogenic emissions. Normally they should be in the same horizontal resolution as the modeling domain. This means for high resolution domains, that the basis of the anthropogenic emissions have to be very accurate. The following paragraph briefly explains the schematics of such a modeling system. The apprach is applied by the Earth Science Group of the Barcelona Supercomputing Center (BSC). Other operational CWF's are listed in Kukkonen et al. (2012).

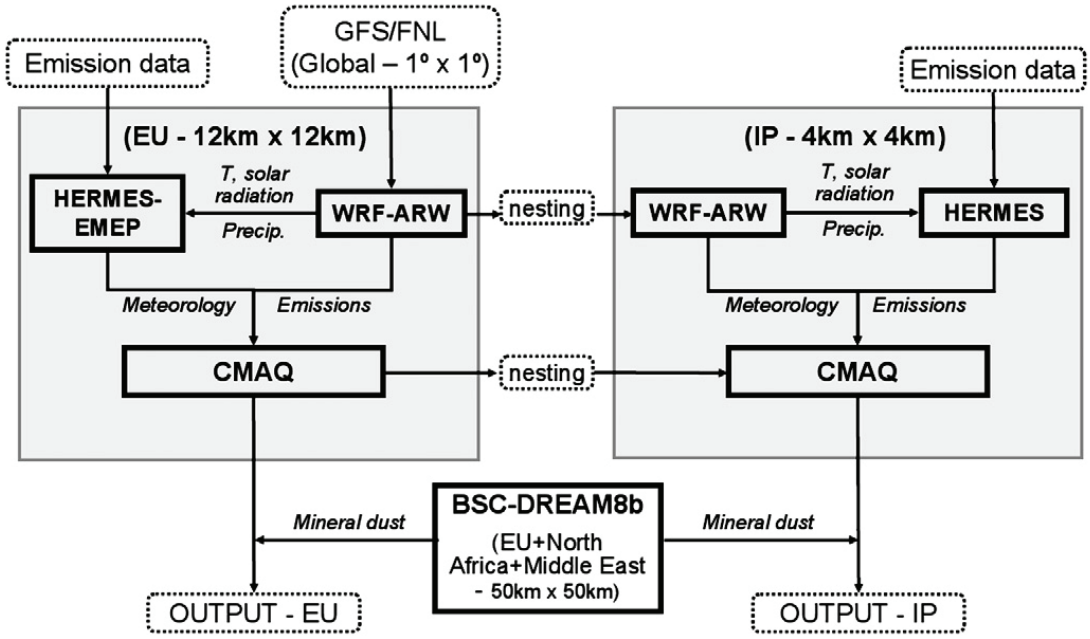

Figure 2: Modular structure of the modeling system used to simulate air quality dynamics in Spain. From Baldasano et al. (2008*b*).

The Earth Science group of the BSC developed a state-of-the-art air pollution modeling system for the Iberian Peninsula. The schematics of the system are shown in Fig. 2. Like CTM, they use the off-line model CMAQ, but also tested the application of the CHIMERE model (Jorba et al., 2008; Baldasano et al., 2008*b*). Overall, an European domain with a horizontal resolution of 12 km and a nested domain for the Iberian Peninsula with a resolution of 4 km. Meteorological input data for the coarser domain are derived from the Global Forecast System (GFS). The WRF model served as NWP model. Anthropogenic emissions are derived from the European Monitoring and Evaluation Program (EMEP) and from a specific emission model for Spain (Baldasano

et al., 2008*a*) called HERMES. Additionally they added a mineral dust model, as Saharan dust dispersion is of great interest especially for parts of Southern Spain (Pérez et al., 2006*a*,*b*). Details of the modeling system and validation of the system can be found in Baldasano et al. (2008*b*, 2011) and Pay et al. (2010). This study tried to apply a similar approach but with adapted components.

1.3.3. Air pollution modeling for Switzerland

There are only a few studies on air pollution modeling for Switzerland. Overall, they can be categorized into three different modeling systems. The first one was developed by a private company (GAIASENS Technologies SARL) and was initially granted by an innovation support from the École Polytechnique Fédéral de Lausanne (Kirchner et al., 2001; Couach et al., 2004). Their Mesoscale Air Pollution 3D modelling (Map3D) consists of an European domain (50 km), a domain for Alpine regions (15 km) and additionally the possibility to calculate high resolution domains. The two coarser domains are calculated by the off-line CHIMERE model with input data from the predecessor of WRF and anthropogenic EMEP emissions. These two domains are actually operational and the results for a three day forecast can be seen at `www.gaiasens.com/app/sites/pollution/9/lang:eng`. The higher spatial resolutions are calculated with the Meteorology and Atmospheric Photochemistry Mesoscale model (MetPhoMod) which was mainly developed by the Geographical Institute of the University of Bern. However, MetPhoMod was never applied to the entirety of Switzerland or for an entire simulated year. Small scale studies were conducted using this approach for areas of Strasbourg and Grenoble (Couach et al., 2002, 2003, 2004).

The second Swiss modeling system was developed by the Paul Scherrer Institute (PSI). The applied CTM model is the off-line model CAMx and is driven by the meteorology of WRF runs (Andreani-Aksoyoglu et al., 2008; Keller et al., 2008; Aksoyoglu et al., 2011). This modeling system was only applied to specific studies and not operationally. All studies so far modeled two domains with 27 and 9 km, respectively. Anthropogenic emissions are from the Netherlands Organization from disaggregated EMEP emissions and high resolution emissions from SAEFL (2003*a*, 2004). The same basis of Swiss federal emissions is used for this study (see Chap. 2.5.1). Overall, both of these air pollution modeling systems have not calculated a Swiss domain for an entire year and both modeling systems use off-line models as CTM's. Due to the first fact, these systems were never used for epidemiological studies so far.

The last approach to assess spatial air pollution concentrations in Switzerland is based on an empirical dispersion model. The PolluMap (version 2.0) model is developed by two private companies (Meteotest and INFRAS) and commissioned by the Federal Office for the Environment (FOEN). The detailed description of PolluMap is elaborated in SAEFL (2003*a*, 2004) and FOEN (2011). PoluMap is an empirical model which uses transfer functions to represent the impact of emission sources on neighboring areas. The

aforementioned emission basis was especially developed for PolluMap (SAEFL, 2003*a*, 2004; FOEN, 2011). The emissions are available for NO_x, PM_{10} and $PM_{2.5}$ with a horizontal grid of 200 m and 100 m for the basis years 2000 and 2010, respectively. The anthropogenic Swiss emissions are further described in Chap. 2.5.1. In addition to the dispersion function based on a Gaussian distribution, PolluMap adds a rural-urban transfer function and spatial background concentrations based on measurements. The model itself simulated the annual mean concentrations of NO_2, PM_{10} and $PM_{2.5}$ for a 200 m resolution. This approach was used in prior SAPALDIA studies (Liu et al., 2007). Chap. A2-4.3 and Fig. A2-8 shows a comparison of the WRF-Chem results of this study to the results of PolluMap.

All these three models have their downsides for estimating population based air pollution exposure assessment. Therefore the decision was taken to apply a different approach to achieve air pollution estimates. An on-line CTM was preferred over a off-line air model as applied by PSI or GAIASENS Technologies SARL. The final decision was to apply WRF-Chem to entire Switzerland with a high horizontal resolution.

1.4. Outline

After this introduction on the motivation of this thesis and the current state of research, the applied methods and used datasets will be explained (see Chap. 2). It consist of a brief description of the applied NWP model and CTM and the chosen options. Then, the model setup with the different domains and the used input datasets are explained. The biggest effort to achieve the goal of this study has to be invested in the development of the high resolution anthropogenic emissions (see Chap. 2.5). Measurements which were used for the validation of the modeling system as well as the statistical parameters are further described. The last two sub-sections explain the needed computational resources and an applied post-processing algorithm. Results are split up into three different parts. The first one is an evaluation of the coarser European domain with focus on different IC/BC's. The results are presented in the form of a peer-reviewed journal article which was accepted by Meteorology and Atmospheric Physics[A1]. The second part is also in form of a journal article (Atmospheric Research)[A2]. It is a validation of the Swiss domain results (years 1991 and 2002) compared to ground-based measurements as well as PolluMap. Results from 2010, spatio-temporal differences to 2002 and diurnal cycles are presented in the third results part. The thesis ends with short summaries of the most important topics, the study limitations and the relevance of this work to the research field of exposure assessment. Finally an outlook for possible future studies and overall conclusive remarks are provided.

A1: Ritter et al. (2012), *Meteorol. Atmos. Phys.* **[in press]**
A2: Ritter et al. (2013), *Atmos. Res.* **[in review]**

2. Method and Data

This chapter provides a short description of the numerical weather prediction model, the chemical transport model and their setup. Initial and boundary conditions (IC/BC) are briefly explained and the formation of spatial emission datasets are illustrated. Used measurements, statistical parameters and computational resources are near the end of this chapter. At the end of this chapter, a post-processing approach with multi-linear regressions is elaborated.

2.1. Numerical weather prediciton model - WRF

The Weather Research and Forecasting (WRF) model version 3.2.1. is used for simulating meteorological parameters. The model was mainly developed by the National Center for Atmospheric Research (NCAR), the National Oceanic and Atmospheric Administration (NOAA), the National Centers for Environmental Prediction (NCEP), the Earth System Research Laboratory, the Department of Defense's Air Force Weather Agency, the Naval Research Laboratory, the Center for Analysis and Prediction of Storms at the University of Oklahoma and the Federal Aviation Administration. It is maintained and supported as a community model and freely available. It is suitable for a broad range of applications such as real-time numerical weather prediction, large-eddy simulations, air quality modeling, regional climate simulations and hurricane simulations. Two different dynamical solvers exist, whereas this study focuses on the Advanced Research WRF solver (ARW) as the Nonhydrostatic Mesoscale Model (NMM) solver is not compatible with the chemistry extension. WRF-ARW is a mesoscale, fully compressible, Eulerian non-hydrostatic model with several options for physical parameterization (Skamarock and Klemp, 2008*b*; Skamarock et al., 2008*a*). Microphysical processes are treated in this study by the Eta grid-scale cloud and precipitation scheme (Eta Ferrier), which predicts water vapor and condensate in cloud water, rain, cloud ice and precipitation ice (Rogers et al., 2001). The sub-grid-scale effects of convective and shallow clouds are handled by the Betts-Miller-Janjic scheme (Janjic, 1994, 2000). Cumulus parameterization is normally only applied to horizontal grid resolutions bigger than 5 km. However, the Betts-Miller-Janjic scheme does not modify sub-cloud layer and has no mass-flux parameterization. It is therefore suitable for any horizontal resolutions (Gilliland and Rowe, 2007). The calculation of friction velocities and exchange coefficients for the land-surface models and the planetary boundary layer scheme are implemented by the Eta surface layer scheme (Janjic, 1996, 2001). It is based on the Monin-Obukhov similarity theory (Monin and Obukhov, 1954). The Noah land-surface scheme is used to provide heat and moisture fluxes over land points and sea-ice points (Chen and Dudhia, 2001). It is a 4-layer soil temperature and moisture model with canopy moisture and snow cover prediction and provides the sensible and latent heat fluxes to the boundary layer scheme. The land use and land cover datasets from the United States Geological Survey (USGS) provided the needed variables for

the Noah land-surface scheme. Vertical sub-grid-scale fluxes caused by eddy transport are treated through the Mellor-Yamada-Janjic planetary boundary layer scheme (Janjic, 1990, 1996, 2001). Atmospheric Radiation is simulated by the Rapid Radiative Transfer Model (RRTM) for longwave (Mlawer et al., 1997) and the Dudhia scheme for shortwave (Dudhia, 1989) radiation, respectively. As overview, the physicals option are listed in Tab. A2-1.

2.2. Chemical transport model - WRF-Chem

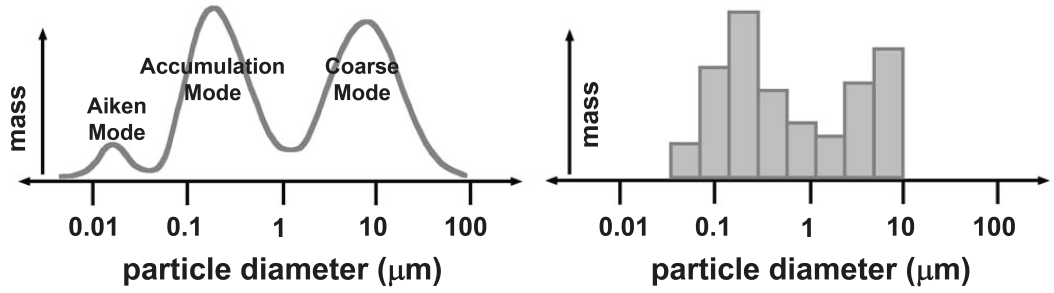

Figure 3: Modal aerosol approach (left side) as used in MADE and a eight sectional aerosol bin approach (right side) as used in MOSAIC. From Fast and Gustafson (2007).

For the simulation of air pollutants the on-line approach using the WRF chemistry extension (WRF-Chem) version 3.2.1 was applied (Grell et al., 2005; Fast et al., 2006). The model is developed by the NOAA, the Pacific Northwest National Laboratory, the Cooperative Institute for Research in Environmental Sciences, the University Corporation for Atmospheric Research (UCAR), NCAR, the Max Planck Institute in Mainz (Germany), the University of Chile and the Centro de Previsao de Tempo e Estudos Climaticos (Brazil). The on-line approach has the advantage of using the same time steps (or a multiple of it), grid cells and transport scheme as WRF and is fully consistent with the meteorological components. The model has several options for the gas-phase chemical mechanisms as well as for the aerosol schemes and the photolysis schemes. This study uses the Carbon bond mechanism version Z (CBM-Z) which has over 65 prognostic species and over 160 chemical reactions (Zaveri and Peters, 1999; Fast et al., 2006; Ritter, 2010). CBM-Z is based on a lumped structure approach that involves lumping organics according to the types of bond present in their molecular structures. Therefore, fewer categories are needed to represent the bond groups which reduces computing time. Aerosols are treated by the corresponding Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) using four sectional bins (Zaveri et al., 2008; Fast et al., 2006; Ritter, 2010). This approach was chosen as a modal design implemented in the Modal Aerosol Dynamics Model for Europe (MADE) has given

unreasonable results over complex terrain. Fig. 3 shows the differences of a sectional approach (right side) and a modal approach (left side). In this case the modal approach would only have to process and internally save three different aerosol groups, whereas the sectional approach depends on how many bins are used (eight in this figure). The different bins within MOSAIC are considered as internally mixed. Slightly better results were achieved with eight sectional bins but considering the additional computation time and the only little gain, the best option is to only take four bins. Aerosols are implemented as primary particles (sea salt, soil dust, black carbon and organic carbon) and secondary particles formed by various salts, inorganic gas-phase chemistry, coagulation and homogeneous nucleation. Secondary organic aerosols (SOA) are only implemented from the WRF-Chem version 3.3 on for MOSAIC and therefore not present in this study. The Fast-J scheme (Wild et al., 2000) calculates the needed photolysis frequencies and dry deposition velocities are calculated using an approach developed by Wesely (1989). Direct and indirect radiative feedback and wet scavenging were not implemented as they were incompatible with some chosen physics options.

2.3. Model setup

The model is set up for a high resolution domain nested into a coarser one. The coarser European domain covers an area from northern Africa to southern Finland. It has a horizontal resolution of 30 km with 27 vertical sigma layers (see Fig. 4) up to 50 hPa. Overall, the domain has 100 by 100 grid points. However, for testing the output of the European domain the study design was changed, so that the model resolution matches the European anthropogenic emissions. Therefore the validation of the European domain was done for exactly the same extended area, but with a horizontal resolution of only 50 km with 60 by 60 grid points. The decision for using a 30 km resolution for the nesting approach is based on a short pre-study with meteorological parameters. The pre-study revealed, that the model sometimes gets instable with a parent-grid ration of 25 (with a horizontal resolution for the Swiss domain with 2 km). On the other hand, no downsides could be seen

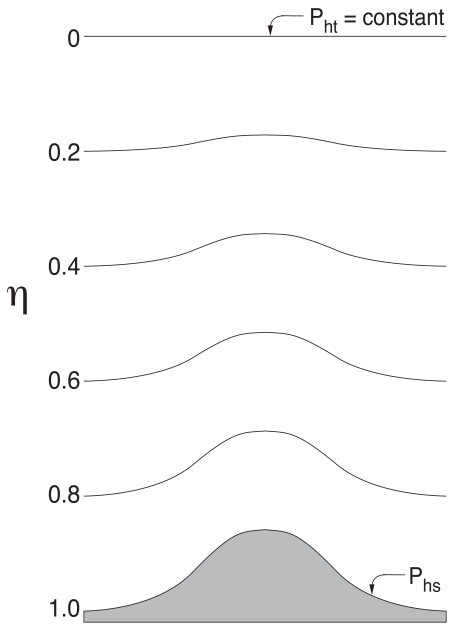

Figure 4: WRF-ARW sigma coordinate. From Skamarock et al. (2008*a*).

for a parent-grid ratio of 15 as results were satisfactory. As chemical parameters are just passed as a scalar field, they do not interfere with the nesting approach. Internal model timesteps are bound to the resolution of the domain. Normally, a factor of six of the resolution in km should be applied in seconds. Following this calculation, the internal timesteps were five and three minutes, respectively. The chemical timesteps can be a multiple of the ones for the meteorological parameters, but were the same for the coarser European domain. Photolysis rates were updated every half hour. The final output is stored into daily files with an hourly resolution. The extend of the coarser European domain can be seen in Fig. 5.

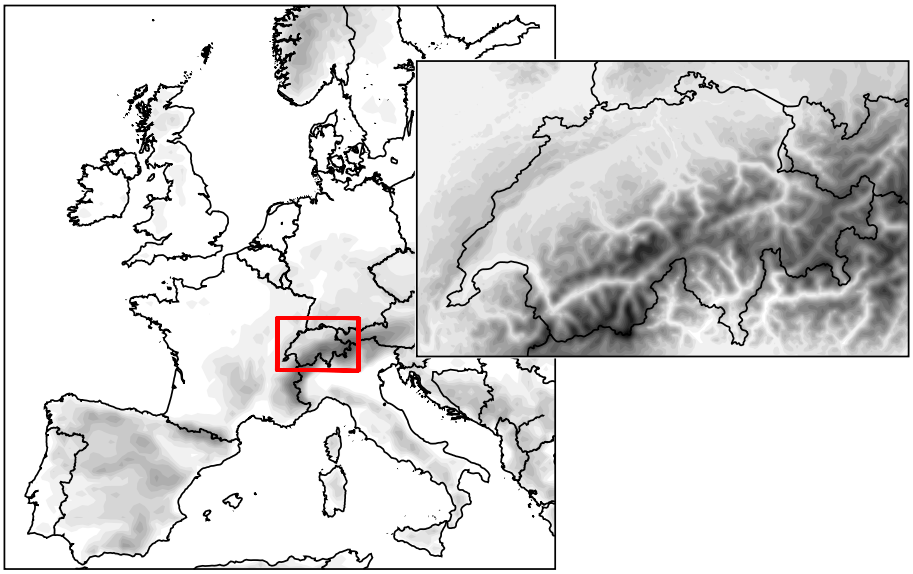

Figure 5: Domain setup of the air pollution modeling system.

For all nesting approaches, the lateral BC's of the high resolution domain are interpolated from the coarser grid. In a 1-way nesting, this is the only information that is exchanged by the model. A 2-way nesting gives the information of the high resolution run back to the coarser grid, so that the grid cells of the coarser domain are replaced by the high resolution information. Both methods have to be run simultaneously. However, the 1-way nesting approach can also be split up into two separate steps. First the coarser domain is calculated and after a special processing step to extract the lateral BC's and also the IC's of the high resolution domain, the latter can be run. The 1-way nesting approach with two separate runs has been chosen for this study for the simplicity to be able to calculate one domain after the other.

The high resolution Swiss domain is set up with a 2 km horizontal resolution and for 27 vertical sigma layers (see Fig. 4). It covers the entirety of Switzerland and has 210 by 135 grid points. The Swiss domain can be seen in Fig. 5. As can be seen, the

Swiss domain is situated in the middle of the coarser domain and the influence of the boundaries of the coarser domain are minimal to Swiss domain. The initial idea would have been to calculate the Swiss domain with a resolution of 1 km. However, this would have let to a computation factor multiplied approximately by ten. Internal timesteps for the meteorological parameters were every twelve seconds and the chemical timesteps were every 36 seconds. The factor between those timesteps was chosen to decrease overall computing time. Photolysis rates were updated every two minutes. As for the European domain, the final results were stored in daily files with an hourly resolution.

Overall, the years 1991, 2002 and 2010 were calculated. This was achieved by simulating 5-day steps. This means that one single run calculated five consequent modeled days. Fig. 6 shows a schematic of the temporal modeling setup. The additional 12 hours prior to the 5-day period is needed, to eliminate effects of the newly initiated meteorological parameters. The blue arrows show the chemical initialization for the consequent runs.

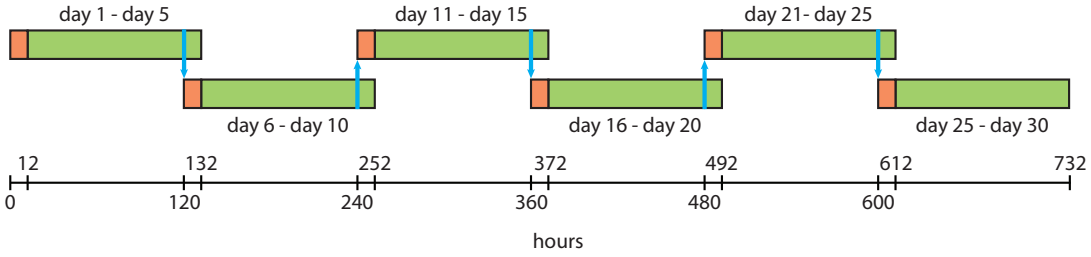

Figure 6: Schematics of the temporal modeling setup.

2.3.1. Topography smoothing

The ARW dynamics core of the WRF model has stability problems when used for very complex terrain (e.g. Switzerland) with a high horizontal resolution. To resolve this problem one option would be to increase diffusion rates. This would smooth every meteorological and chemical variable. A better solution is to smooth the actual terrain information prior to pass it to the model. The high resolution topography data (USGS 30s) was smoothed with a hardcoded smooth-desmooth algorithm from the WRF Preprocessing System (WPS). This algorithm had to be iterated four times to have the model behave stably. However, as this study did not want to loose the topography information in lower regions, where most of the people live in Switzerland, the algorithm was only applied for pixels with an altitude higher than 1,000 m. Fig. 7 shows the applied smooth-desmooth algorithm. The left side shows the initial field with the nine inner pixels of values of 9.0 (green). All other pixels have values of 1.0. After applying the algorithm four times, the regions with decreased (red) and increased (green) values

can be seen. The center pixel is even higher at the end with a value of 9.37. The smoothed topography for pixels with an altitude over 1,000 m can be seen in Fig. A2-1 and Fig. 5.

before smoothing

1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
1.00	1.00	1.00	1.00	1.00	9.00	9.00	9.00	1.00	1.00	1.00	1.00	1.00
1.00	1.00	1.00	1.00	1.00	9.00	9.00	9.00	1.00	1.00	1.00	1.00	1.00
1.00	1.00	1.00	1.00	1.00	9.00	9.00	9.00	1.00	1.00	1.00	1.00	1.00
1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00

smooth=smth-desmth; smooth_pass=4

1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
1.00	1.00	1.00	1.00	0.98	0.94	0.92	0.94	0.98	1.00	1.00	1.00	1.00
1.00	1.00	1.02	1.01	0.88	0.67	0.57	0.67	0.88	1.01	1.02	1.00	1.00
1.00	1.00	1.01	1.00	0.96	0.90	0.87	0.90	0.96	1.00	1.01	1.00	1.00
1.00	0.98	0.88	0.96	1.66	2.78	3.35	2.78	1.66	0.96	0.88	0.98	1.00
1.00	0.94	0.67	0.90	2.78	5.79	7.34	5.79	2.78	0.90	0.67	0.94	1.00
1.00	0.92	0.57	0.87	3.35	7.34	9.37	7.34	3.35	0.87	0.57	0.92	1.00
1.00	0.94	0.67	0.90	2.75	5.79	7.34	5.79	2.75	0.90	0.67	0.94	1.00
1.00	0.98	0.88	0.96	1.66	2.78	3.35	2.78	1.66	0.96	0.88	0.98	1.00
1.00	1.00	1.01	1.00	0.96	0.90	0.87	0.90	0.96	1.00	1.01	1.00	1.00
1.00	1.00	1.02	1.01	0.88	0.67	0.57	0.67	0.88	1.01	1.02	1.00	1.00
1.00	1.00	1.00	1.00	0.98	0.94	0.95	0.94	0.98	1.00	1.00	1.00	1.00
1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00

Figure 7: Smooth-Desmooth algorithm (four iterations) with an initial field of nine inner pixels with higher values.

2.4. Boundary and initial conditions

Initial conditions (IC) are needed to have the initial spatial information of the domain. Lateral boundary conditions (BC) adds the information which is coming from the boarders of the domain into it. Therefore the IC is only a time-snap of the whole domain, whereas the BC's are 2-dimensional fields for every border. The latter information has to have a temporal resolution. IC/BC's for the Swiss domain are coming from the coarser European domain. BC's are updated every hour for all meteorological and chemical variables. For the European domain an evaluation study was made for different chemical BC's as well as different meteorological IC/BC's. Results can be seen in Chap. 3.1. These tests were carried out for July 2005, because data of one meteorological IC/BC does not exist for the first two modeled years (1991 and 2002). Chemical IC's are not needed when an additional spin-up calculations of five days precedes, as the chemical fields are mainly driven by the emissions. After these first five days the chemical IC's are taken from the preceding runs (see Fig. 6).

2.4.1. Chemical boundary conditions

The WRF modeling system has hardcoded chemical BC's. They are based on an idealized, northern hemispheric, mid-latitude, clean environmental, vertical profile from the NOAA Aeronomy Lab Regional Oxidant Model (NALROM). A description of the NALROM model can be found in McKeen et al. (1991), Liu et al. (1996) and Peckham et al. (2010). There are no spatial and temporal differences as the vertical profile is declared globally and has no temporal resolution.

The other chemical BC's are derived from the global climate model from the Laboratoire de Météorologie Dynamique (LMDZ) with the Interaction with Chemistry and Aerosols (INCA) model (Hauglustaine et al., 2004; Szopa et al., 2009). The used data are monthly values of the model for the mean of the years 1997-2001. The model has a horizontal resolution of 3.75 degrees by 2.5 degrees and 19 vertical sigma levels (up to 3 hPa). The interpolation between the LMDZ-INCA grid and the used grid in WRF was done by a weighted mean of the surrounding eight grid points (3D) of the LMDZ-INCA model. The interpolation method is shown in Fig. 8. This graphs shows the European domain with a horizontal resolution of 50 km (60 by 60 grid points). This procedure could only be applied for eight different chemical species (O_3, NO, NO_2, HNO_3, PAN, H_2O_2, CO, HCHO) as others are not implemented in the LMDZ-INCA model. For all other needed chemical species the values from the NALROM vertical profile were applied as chemical BC.

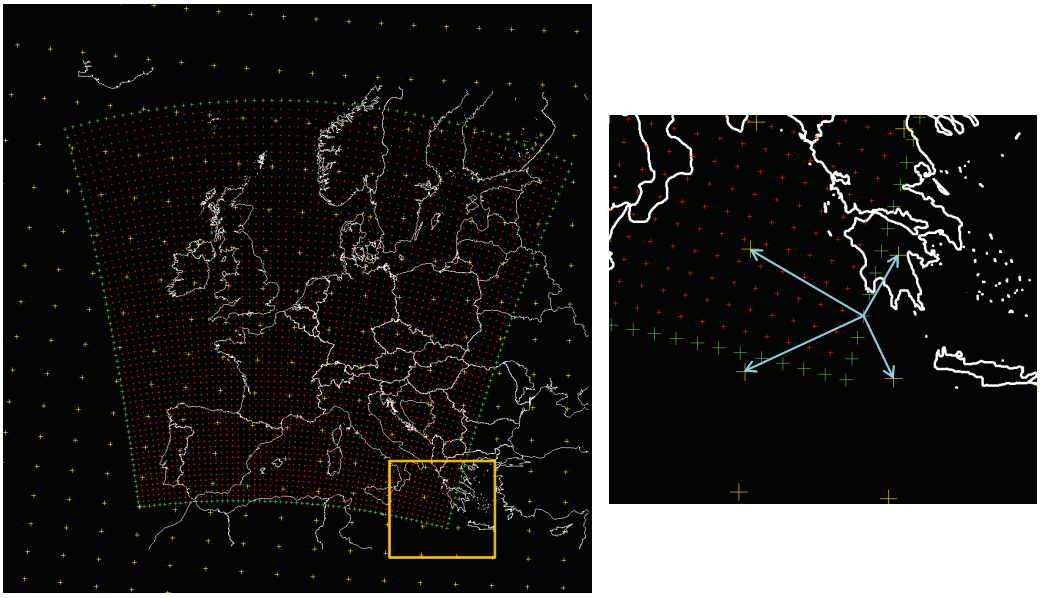

Figure 8: Interpolation method between the LMDZ-INCA model (yellow crosses) and the European domain (red crosses). The green crosses are the boundary pixels of the European domain (50 km resolution). The interpolation was done with a weighted mean for all three dimensions.

2.4.2. Meteorological initial and boundary conditions

Two different global models served as meteorological IC/BC's. The first one is the GFS which is available from NOAA. The horizontal resolution is 1.0 degree (original spectral grid: T382L64) for July 2005 and for 2010. No GFS data is available for the years

1991 or 2002. This was also the reason for testing the effect of different meteorological IC/BC's on air pollutants for the European domain. Only the initial and first forecast step for every 6-h GFS run was taken so that a temporal resolution of 3 hours finally existed.

The second model is Reanalysis data from NCEP. These datasets are continually gridded and incorporate observations and numerical weather prediction model output. They are specially made for reanalysis of past years and date back to 1948. However, revised Reanalysis data (Reanalysis II) are available from 1979 onwards. Therefore, the Reanalysis II datasets can fill the lack of GFS availability for the years 1991 and 2002. Reanalysis II data have a horizontal resolution of 2.5 degree (original spectral grid: T64L28) and a temporal resolution of 6 hours (Kanamitsu et al., 2002). The main differences of GFS and Reanalysis II data are the temporal resolution and more important the spatial resolution.

2.5. Emissions

For both domains, spatio-temporal emissions are needed. At least anthropogenic and biogenic emissions have to be distinguished. Additionally dust and sea salt emissions can play an important role depending on the chosen domain. Anthropogenic emissions are by far the most important emissions. There are emissions inventories for macro-scale regions (e.g. Europe, U.S.A) with rather low horizontal resolution. However, anthropogenic emissions have special characteristics for every domain. For this reason local data has to be implemented and the emissions can not be modeled by a global emission model for high resolutions. The overall modeling results depend strongly on very accurate high resolution anthropogenic emissions.

2.5.1. Anthropogenic emissions

Anthropogenic emissions for the European domain are taken from the European Monitoring and Evaluation Program (EMEP). These gap-filled, anthropogenic emissions have a horizontal resolution of 50 km (Vestreng and Klein, 2002; EEA, 2009). EMEP emissions consists of yearly averages for five main pollutants, heavy metals, persistent organic pollutants and particulate matter for eleven different SNAP sectors (Selected Nomenclature for reporting of Air Pollutants). The different SNAP sectors are listed in Tab. 2. For this study, only the main pollutants (CO, NH_3, NMVOC, NO_x, SO_x) and the particulate matter ($PM_{2.5}$, PM_{coarse}) were needed. Afterwards different disaggregation, conversion and transformation steps have to be applied. A schematic summary of the processes are shown in Fig. 9. The different processes are elaborated in the following paragraphs.

Table 2: Selected Nomenclature for reporting of Air Pollutants (SNAP) sectors.

No.	Sources
SNAP 1	Combustion in energy and transformation industries
SNAP 2	Non-industrial combustion plants
SNAP 3	Combustion in manufacturing industry
SNAP 4	Production processes
SNAP 5	Extraction and distribution of fossil fuels and geothermal energy
SNAP 6	Solvent use and other product use
SNAP 7	Road transport
SNAP 8	Other mobile sources and machinery
SNAP 9	Waste treatment and disposal
SNAP 10	Agriculture
SNAP 11	Other sources and sinks

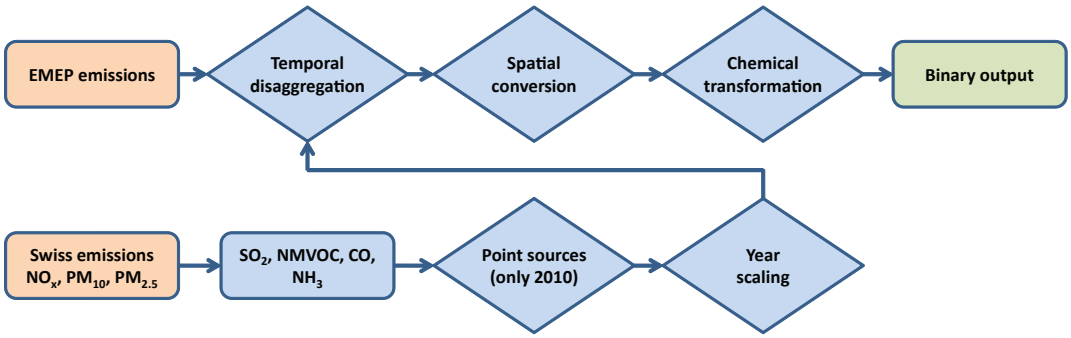

Figure 9: Schematics of the processing steps for the anthropogenic emissions.

The anthropogenic emissions for the Swiss domain are based on federal emissions obtained from SAEFL (2003*a*, 2004) and FOEN (2011). These emissions exist for NO_x, PM_{10} and $PM_{2.5}$, whereas the latter is a fraction of the values of PM_{10}. The annual emission map of NO_x is shown in Fig. 10. All three pollutants are available for the reference years 2000 and 2010. The horizontal resolution for the emissions of the year 2000 and 2010 are 200 m and 100 m, respectively. They are split up into different categories as residential, industry & commerce, agriculture & forestry and different transportation modes (road, rail, water, air). Most of these emission-categories are built with a top-down approach. The missing emission datasets for SO_2, NMVOC, CO and NH_3 had to be developed especially for this study. The same top-down approach has been applied for these emissions as the one for NO_x. The Swiss total amount per subcategory has been adapted with the help of various federal publications (SAEFL, 1995, 2004; FOEN, 2008, 2010*a*) as well as the emission values delivered to the United Nations

Economic Commission for Europe (UNECE) Convention on Long-range Transboundary Air Pollution (CLRTAP) (FOEN, 2010*b*). For 2010, point sources were implemented with data from the SwissPRTR (Swiss Pollutant Release and Transfer Register). As the emission reference year (2000) did not match the first two modeled years (1991 and 2002), the spatial emission-categories were scaled to Swiss total amounts delivered to the UNECE-CLRTAP. This means that the emissions for 1991 and 2002 have the same base year (2000), whereas the emissions for 2010 have their own reference year. All emissions could be reallocated into the eleven SNAP sectors according to detailed sub-categories. The remaining steps are the same as the one for the EMEP emissions (see Fig. 9).

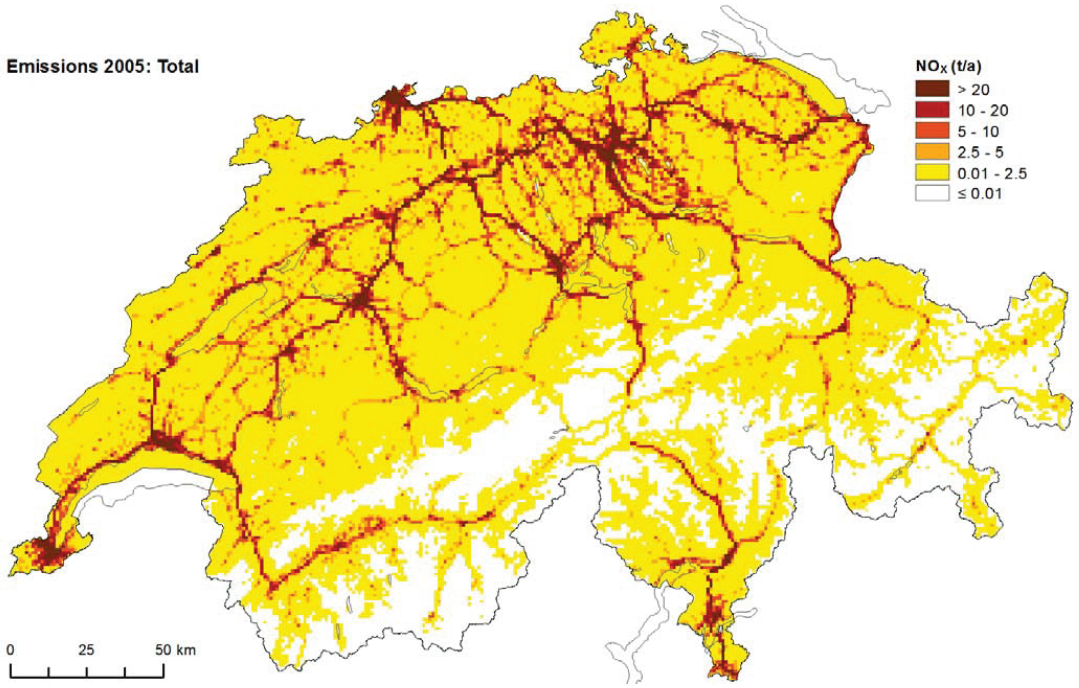

Figure 10: Total amount of NO_x anthropogenic emissions in Switzerland for 2005. Plotted on a grid mesh size of 1 km. From FOEN (2011).

Temporal disaggregation The annual emissions were temporally disaggregated into hourly emissions according to the SNAP sectors, the country codes and the time factors from the Generation and Evaluation of Emission Data (GENEMIS) program (Friedrich and Reis, 2004; Tuccella et al., 2012). The formulas for the disaggregation from yearly to monthly, daily and hourly emissions are shown below. *Emiss* stands for the emissions, *CorrFac* for the correction factors and d for the day of the week.

$$Monthly\,Emiss = \frac{Annual\,Emiss}{12} \cdot Monthly\,CorrFac$$

$$Daily\,Emiss = Monthly\,Emiss \cdot \frac{Daily\,CorrFac}{\sum_{d=1}^{7} Daily\,CorrFac_d \cdot \#\,of\,days\,per\,month_d}$$

$$Hourly\,Emiss = \frac{Daily\,Emiss}{24} \cdot Hourly\,CorrFac$$

Spatial conversion The spatial transformation from the EMEP and Swiss emission projections to the WRF projections were accomplished with the inverse next neighbor interpolation. The interpolation scheme is shown in Fig. 11. For EMEP emissions the 50 by 50 km tiles were split into 5 by 5 km tiles before being reallocated. The ground-based emissions were also vertically disaggregated to the lowest six sigma layers of the WRF modeling system according to the factors used in the unified EMEP model per SNAP sector.

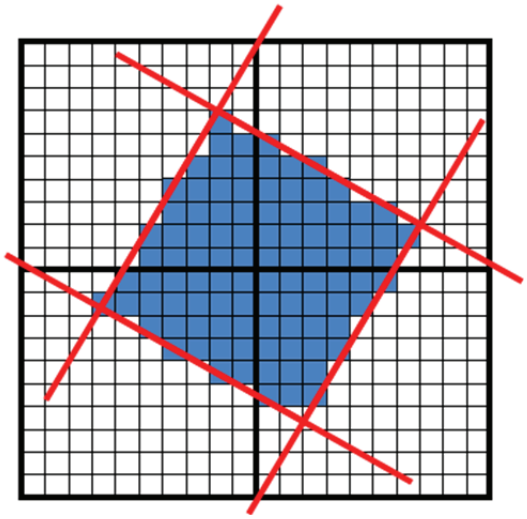

Figure 11: Inverse next neighbor interpolation. The red lines are the desired grid and the thick black lines are the available grid. The thin black lines mark the subgrids which will be reallocated to the desired grid (blue rectangles). From Ritter (2009).

Chemical transformation The last conversion step consists of adapting the chemical species system from the five main pollutants and the particulate matter to the needed CBM-Z/MOSAIC input. This conversion was done in different steps. The first one was a conversion from the EMEP scheme to the Carbon-Bond mechanism version 4 (CBM4) scheme according to conversion factors (CF) developed by the Earth Science Group of the Barcelona Supercomputer Center (EPA, 2010; Parra, 2004; Arévalo, 2005). The CF's and their explanations are listed in App. A. The CBM4 scheme is an older version of the CBM-Z scheme and most of the variables could directly be used from the CBM4 scheme. A summary of the conversion between the different schemes are shown in Tab. 3. The CF's from the CBM4 to the CBM-Z scheme are from Zaveri and Peters (1999) for all NMVOC's and from an WRF-Chem internal routine for splitting up $PM_{2.5}$ emissions into Aitken and accumulation mode.

2.5.2. Biogenic emissions

Biogenic emissions are built up on runtime using the model of emissions of gases and aerosols from nature (MEGAN) version 2.0.4 (Guenther et al., 2006; Sakulyanontvittaya et al., 2008). A slightly simplified version of the standalone model is implemented in the WRF-Chem source code. As there is no explicit canopy model in the WRF code, no soil moisture factors and no production nor loss of emission within the plant canopy are applied. The on-line implementation has the advantage that almost no additional data has to be prepared. All emission factors are based on the functional types, except for isoprene emissions where a separate map of emission factors is used. Overall, the implementation of biogenic emissions from MEGAN does not need much input from the user.

2.5.3. Dust and sea salt emissions

Dust emissions originating from arid regions (e.g. Sahara) are transported over long periods (Zhao et al., 2010; Kang et al., 2011). This is an important factor for the southern regions of Europe (e.g. Spain). To take this fact into account the hardcoded MOSAIC dust emissions option was applied. These dust emissions are built up from several datasets that are already used by WRF (e.g. USGS datasets). The impact of dust emissions on the Swiss domain is minor, so that manually added more refined dust emissions are not needed. For the same reason manually added more refined sea salt emissions were left out. However, the hardcoded MOSAIC sea salt emissions option was applied (Saide et al., 2012; Yang et al., 2012).

Table 3: Chemical conversion steps of the emissions. Conversion factors from the EMEP scheme to the CBM4 scheme are listed in App. A. Conversion Factors (CF) from CBM4 to CBM-Z are listed in 4‘*th* column.

EMEP	**CBM4**	**CBM-Z**	**CF**	**Species**
CO	CO	co	1.000	Carbon monoxide
NH_3	NH3	nh3	1.000	Ammonia
SO_x	SO2	so2	1.000	Sulfur dioxide
NO_x	NO	no	1.000	Nitric oxide
	NO2	no2	1.000	Nitrogen dioxide
NMVOC	FORM	hcho	1.000	Formaldehyde
	PAR	eth	0.019	Ethane
		hc3	0.079	Alkane
		hc5	0.052	Alkane
		hc8	0.037	Alkane
	TOL	tol	1.000	Toluene
	ALD2	ald	0.116	Acetaldehyde
		ket	0.072	Acetone
	ETH	ol2	1.000	Ethene
	OLE	olt	1.000	Terminal olefin carbons
		oli	1.000	Internal olefin carbons
	XYL	xyl	1.000	Xylene
	ISOP	iso	1.000	Isoprene
PM_{coarse}	PSO4C	so4c	1.000	PM_{coarse} (sulfates)
	PNO3C	no3c	1.000	PM_{coarse} (nitrates)
	POAC	orgc	1.000	PM_{coarse} (organic carbon)
	PECC	ecc	1.000	PM_{coarse} (elemental carbon)
$PM_{2.5}$	PMFINE	pm25i	0.200	$PM_{2.5}$ (unspeciated, Aitken mode)
		pm25j	0.800	$PM_{2.5}$ (unspeciated, accum. mode)
	PSO4	so4i	0.200	$PM_{2.5}$ (sulfates, Aitken mode)
		so4j	0.800	$PM_{2.5}$ (sulfates, accum. mode)
	PNO3	no3i	0.200	$PM_{2.5}$ (nitrates, Aitken mode)
		no3j	0.800	$PM_{2.5}$ (nitrates, accum. mode)
	POA	orgi	0.200	$PM_{2.5}$ (organic carbon, Aitken mode)
		orgj	0.800	$PM_{2.5}$ (organic carbon, accum. mode)
	PEC	eci	0.200	$PM_{2.5}$ (elemental carbon, Aitken mode)
		ecj	0.800	$PM_{2.5}$ (elemental carbon, accum. mode)

2.6. Measurements

2.6.1. Air pollution measurements - European domain

For the European domain, the model validation was done with data from the EMEP Measurement Network (EMEP, 2010) for July 2005. The validation was done for O_3, NO_2, PM_{10}, $PM_{2.5}$ and SO_2, where the latter two were of minor importance. On one hand because there were only few $PM_{2.5}$ measurements done and on the other hand because SO_2 concentrations are very low for the European domain. All ground-based EMEP stations with valid data for July 2005 are shown in Fig. A1-1. All measurements are available in hourly resolution. $PM_{2.5}$ measurements were done at the same stations where PM_{10} was measured. All EMEP stations are classified as rural and additionally only stations with a fixed minimum distance to the boundaries were considered to eliminate artifacts from BC's. At the end, around 100 O_3, 30 NO_2 and PM_{10} and only 17 $PM_{2.5}$ measurements could be used.

2.6.2. Air pollution measurements - Swiss domain

For the Swiss domain, measurements are available in hourly resolution for the years 1991, 2002 and 2010. The ground-based measurements for validating the model are available through the National Air Pollution Monitoring Network (NABEL). Overall, there exist 16 NABEL stations (SAEFL, 2003*b*; EMPA, 2011). Further measurements are available through several cantonal and regional air quality authorities and are provided by the Swiss Society of the responsible for air pollution control (Cercl'Air). The stations are classified into four different categories. All urban, sub-urban and rural stations are used for the validation. The two mountainous stations (Jungfraujoch, Arosa) are not used due to their limited ability to represent an area of 2 by 2 km^2. The stations that were directly beside a freeway were reclassified into a own freeway category. Finally, four different categories (rural, sub-urban, urban, freeway) existed. The validation was done for 1991 and 2002 and is explained in Chap. 3.2. Measured pollutants of interest were O_3, NO_2 and PM_{10}. However, not all stations measured every pollutant throughout these two years (e.g. almost no measurements for PM_{10} in 1991). Fig. A2-1 shows the stations with valid data for at least one coherent month.

2.6.3. Meteorological measurements

For the Swiss domain (years 1991 and 2002) a short validation for some meteorological parameters were done. The focus was mainly on temperature (2 m above ground) and solar radiation, as these two parameters can have a big influence on air pollutants. Measurements are done by the Swiss Federal Office of Meteorology and Climatology

within their automatic Swiss measurement network (MeteoSwiss, 2004). The stations are uniformly distributed over Switzerland. There were 68 temperature measurements for 1991 and 87 for 2002. For solar radiation there were 65 and 67 stations for 1991 and 2002, respectively.

2.7. Statistical parameters

This study focused on the mean bias (MB), the mean absolute gross error (MAGE), the root mean square error (RMSE) and the Pearson correlation coefficient (R). These parameters are suggested in various scientific literature (Seinfeld and Pandis, 2006; Jacobson, 2005; de Meij et al., 2009). The formulas below show the equations with *OBS* being the observations and *MOD* their corresponding modeled grid points. The number of stations is represented by m and each station has n measurements.

$$MB = \frac{\sum_{i=1}^{n}\sum_{j=1}^{m} MOD_{i,j} - OBS_{i,j}}{nm}$$

$$MAGE = \frac{\sum_{i=1}^{n}\sum_{j=1}^{m} |MOD_{i,j} - OBS_{i,j}|}{nm}$$

$$RMSE = \sqrt{\frac{\sum_{i=1}^{n}\sum_{j=1}^{m} (MOD_{i,j} - OBS_{i,j})^2}{nm}}$$

$$R = \frac{\sum_{i=1}^{n}\sum_{j=1}^{m} (MOD_{i,j} - \overline{MOD}_{i,j})(OBS_{i,j} - \overline{OBS}_{i,j})}{\sqrt{\sum_{i=1}^{n}\sum_{j=1}^{m} (MOD_{i,j} - \overline{MOD}_{i,j})^2}\sqrt{\sum_{i=1}^{n}\sum_{j=1}^{m} (OBS_{i,j} - \overline{OBS}_{i,j})^2}}$$

2.8. Computational resources and architecture

All kinds of chemical transport model (CTM) with a physical-chemical calculation of air pollutants use a lot of computational resources. The used resources are dependent on the hardware architecture, the simulated domain and the chosen options of the model. Only clusters are capable of fulfilling the requirements satisfactorily. The development of the hardware components and the infrastructure has rapidly increased over the last decades (see Fig. 12). However, the architecture of the different chemical transport models have their limitation concerning optimization of computing time. Therefore, such applications still need a lot of computing time.

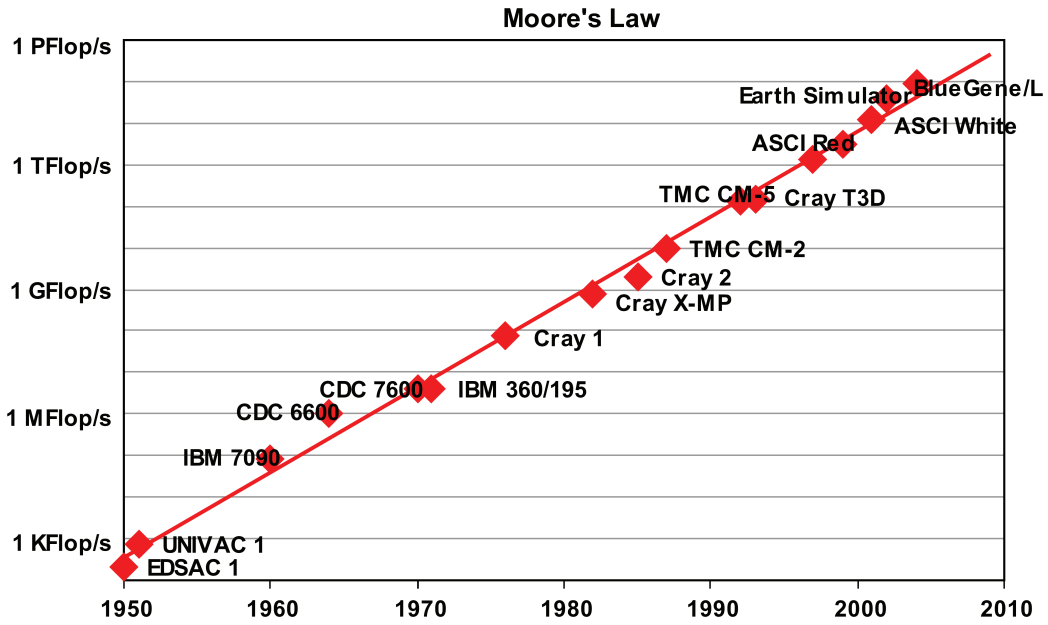

Figure 12: Performance of the fastest computer systems for the last six decades (Moore's Law). From Strohmaier et al. (2005).

All final results for this project were calculated on the cluster of the University of Basel. The cluster has over 80 Intel Nehalem Quad Core (2.66 GHz) processors with 24 GB Memory per two Central Processing Units (CPU)'s and are connected through Infiniband. WRF has the Message Passing Interface (MPI) implemented for parallel computing. The European domain was calculated on 26 cores and the Swiss domain with 50 cores. In real-time the simulation of the European domain needs a little bit less than 2 hours for the calculation of five modeled days whereas the simulation of the Swiss domain needs around 19 hours. Overall, the simulation of five modeled days with all the needed steps endured approximately 24 hours. In an early stage, some tests for the European domain were calculated on the Barcelona Supercomputing Center (BSC).

The model setup scheme for both domains can be seen in Fig. 13. The input datasets have already been explained. The WRF-Chem model consists of the WPS, an initialization routine for real datasets, the meteorological model and the chemistry extension. The model output is in the Network Common Data Form (NetCDF). For manipulating these huge outputs, some smaller routines have been developed for this study. Other steps of the post-processing are a multi-linear regression as well as visualization routines and statistical calculations. At the end, Geotiffs and Shape-Files are automatically generated for all averaged months and years for CO, HCHO, HNO_3, HONO, NO, NO_2, NO_3, O_3, PM_{10}, $PM_{2.5}$ and SO_2.

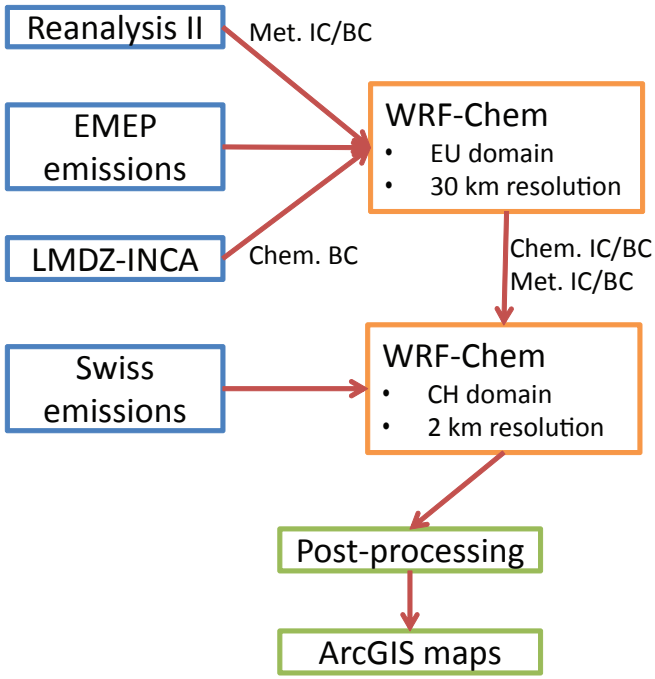

Figure 13: Model setup scheme for both domains.

2.9. Post-processing / multi-linear regression

All kind of models are just a mapping of the reality and therefore introduce a systematic bias. Air quality models are no exception. The biggest sources of systematic bias for CTM's are the limitations of the physical and chemical mechanisms, their interaction and particularly their reliability on emissions. WRF-Chem showed overall too high values for O_3 and too low values for NO_2 compared to measurements. The raw model output of PM_{10} was nevertheless in an acceptable range. However, the observed systematic bias had a seasonal effect and varied by type of location (rural, sub-urban, urban, freeway) of the measurement station. The decision to apply a post-processing routine was taken. The importance of such routines for air quality modeling was already shown by Sicardi et al. (2012), Borrego et al. (2011) and Djalalova et al. (2010). Operational numerical weather prediction often apply post-processing routines. They range from simple methods as bias-correction to more sophisticated methods as Kalman-Filtering or Model Output Statistics (MOS). For this study, several different rather simple methods were tested. On one hand this was the application of simple bias-correction values and on the other hand a multi-linear regression. Predictors for the multi-linear regression were chosen in prior tests by a forward stepwise selection. All predictors can be seen in Tab. 4. The climatological functions are calculated with the day of the year (including fractions for hourly separation) and a conversion factor

for converting degrees to radians. The curves of the climatological predictors are shown in Fig. 14. Both methods (bias-correction and multi-linear regression) were applied for the total of all stations. However, the model was just trained with 50 % of all stations. The other half of the measurements served for the validation of the post-processing routines. The station for the validation was selected randomly if they were not too close to each other. In general for all stations, better results are achieved and the seasonal bias was more or less eliminated. However, a look at the stations classified into the different categories still revealed a systematic category bias. Therefore, both methods were calculated for all categories (rural, sub-urban, urban, freeway) separately. Best results were finally achieved for the multi-linear regression applied for each category. However, the difference between the different post-processing routines are minor.

Table 4: Predictors of the multi-linear regression.

Model	The raw model output
T2	Temperature 2 m above ground
SWDOWN	Solar radiation
PBLH	Planetary boundary layer height
U10/V10	Wind vectors 10 m above ground
Q2	Humidity 2 m above ground
$\sin(DOY)$	Climatological function (red line in Fig. 14)
$\cos(DOY)$	Climatological function (blue line in Fig. 14)
$\sin(0.5 \cdot DOY)$	Climatological function (gray line in Fig. 14)
$\cos(0.5 \cdot DOY)$	Climatological function (black line in Fig. 14)

DOY: day of the year

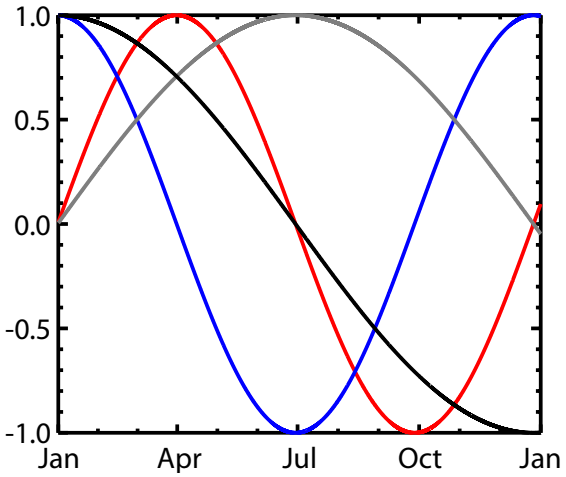

Figure 14: Climatological predictors for the multi-linear regression.

Fig. 15 shows the RMSE for NO_2 for the year 2002 for the raw model output and the different tested post-processing routines. All stations are sorted according the RMSE of the raw model output. Stations without a gray background served for the validation and were not taken into account for the calculation of the post-processing routines. It can be seen that the improvements are significant for over 90 % of the stations, no matter if they served as validation or for calculating the post-processing routines. The improvements were most significant for NO_2, NO and O_3, but still noticeable for PM_{10}. Post-processing routines could not be applied for $PM_{2.5}$ for all modeled years as well as for PM_{10} and NO for the year 1991 due to the lack of sufficient valid measurements.

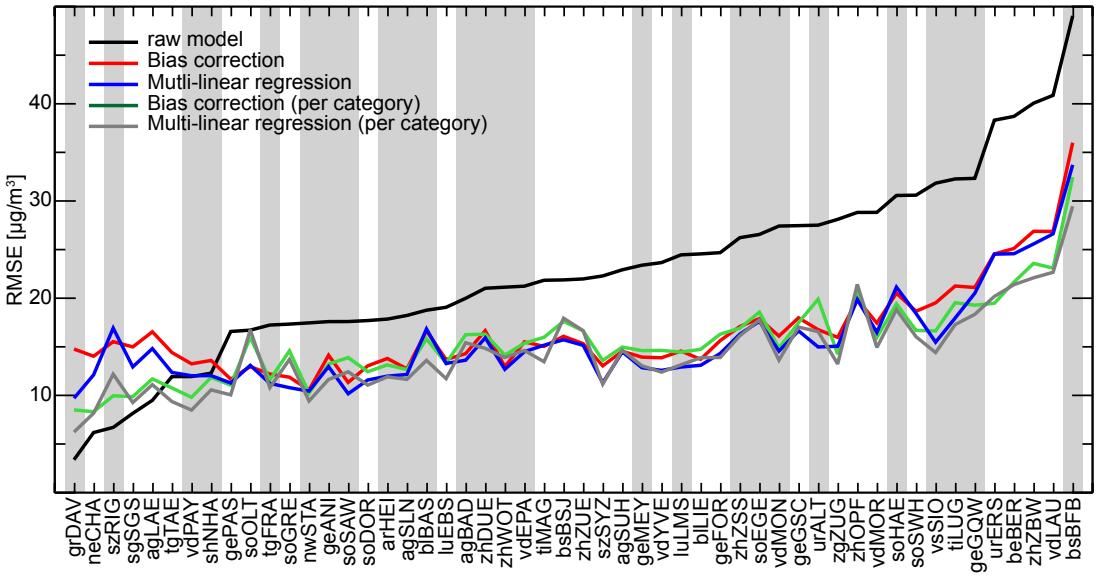

Figure 15: NO_2 root mean square errors for the raw model output and post-processed output (year: 2002). Stations without a gray background served for validation.

Fig. A2-2 is the same figure as Fig. 15 but shows only the raw model output and the multi-linear regression per category. The RMSE between the European domain results and the Swiss measurements were added in Fig. A2-2. Overall, the Swiss domain performs slightly better than the coarser European domain. However, if the multi-linear regression is trained with chemical raw model output from the European domain and meteorological parameters from the Swiss domain, almost the same results are achieved. Conclusions are drawn in Chap. 3.2 as well as in Chap. 4.

The aim of the post-processing routines is to finally have the best available result for the Swiss domain. The influence of the stations that served as input was therefore evaluated. Fig. A2-3 shows the absolute differences of NO_2 RMSE values of the raw model output and the NO_2 RMSE values of the post-processed results. Stations

with gray background are the same as mentioned above and served for calculating the correlation factors for the multi-linear regression. If the selection of the stations would have had a significant influence, a clear pattern could be seen with grayed stations on the left (small absolute differences) and stations with white background on the right (big absolute differences). However, no clear pattern can be distinguished. All available stations with valid data are therefore used for the post-processing routines. All further results (beside if they are explicitly declared as raw model output) are post-processed by a multi-linear regression per location category and trained with all available measurements.

3. Results

The results are shown in three sections. The first one is an evaluation of the European domain with measurements. A special focus was set on chemical boundary conditions (BC) as well as meteorological initial and boundary conditions (IC/BC). This part of the evaluation is from a peer-reviewed article, which was accepted by the Meteorology and Atmospheric Physics journal. The second section is also presented as an article submitted to Atmospheric Research. It covers a validation of the Swiss results for the modeled years 2002 and 1991. The third and last section contains further analysis of Swiss results with a focus on spatio-temporal differences between the years 2010 and 2002 and diurnal cycles.

3.1. European domain

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Meteorology and Atmospheric Physics [**in press**], DOI: 10.1007/s00703-012-0222-8

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ORIGINAL PAPER

Impact of chemical and meteorological boundary and initial conditions on air quality modeling: WRF-Chem sensitivity evaluation for a European domain

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Abstract This study evaluates the impact of different chemical and meteorological boundary and initial conditions on the state-of-the-art Weather Research and Forecasting (WRF) model with its chemistry extension (WRF-Chem). The evaluation is done for July 2005 with 50 km horizontal resolution. The effect of monthly mean chemical boundary conditions derived from the chemical transport model LMDZ-INCA on WRF-Chem is evaluated against the effect of the preset idealized profiles. Likewise, the impact of different meteorological initial and boundary conditions (GFS and Reanalysis II) on the model is evaluated. Pearson correlation coefficient between these different runs range from 0.96 to 1.00. Exceptions exists for chemical boundary conditions on ozone and for meteorological boundary conditions on PM_{10}, where coefficients of 0.90 were obtained. Best results were achieved with boundary and initial conditions from LMDZ-INCA and GFS. Overall, the European simulations show encouraging results for observed air pollutant, with ozone being the most and PM_{10} being the least satisfying.

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1 Introduction

The significance of air pollution modeling is increasing in the field of health assessment. More and more public health cohort studies are using output of air quality models (Liu et al. 2007; Cohen et al. 2009; Bellander et al. 2001), as measurements are expensive and represent only a limited area (especially in mountainous areas). There exists a variety of models to achieve valid results. One of them is the physical and chemical modeling of meteorology and air pollutants. Most of these models are offline meaning that the meteorology is calculated prior to the chemistry in two separate models. A widely used offline model for Europe is the CHIMERE model (Schmidt et al. 2001; Bessagnet et al. 2004; van Loon et al. 2004, 2007; Vautard et al. 2005; Baldasano et al. 2008; Szopa et al. 2009; Pay et al. 2010). Other often used models are the CMAQ (van Loon et al. 2004; Baldasano et al. 2008; Matthias 2008) (especially over America) and the Unified EMEP model (van Loon et al. 2004, 2007). Nevertheless, in this study an online coupled model is used where every grid point and timestep is consistent with the meteorological model. The state-of-the-art Weather Research and Forecasting (WRF) model (Skamarock et al. 2008) with a chemistry extension

(WRF-Chem) (Grell et al. 2005) is used to simulate air pollutants. The approach of adding a chemistry module to an already existing meteorological model is applied to a variety of different meteorological models. Examples beside WRF-Chem are Enviro-HIRLAM (Korsholm et al. 2008), GEM-AQ (Kaminski et al. 2008), COSMO-ART (Vogel et al. 2009), Meso-NH-C (Tulet et al. 2003) and MCCM (Grell et al. 2000).

The overall aim of this study is to calculate concentrations of air pollutants in Switzerland for the years 1991 and 2002 where health surveys of the biggest Swiss cohort study on air pollution and lung diseases in adults (SAPALDIA) were carried out (Ackermann-Liebrich et al. 1997, 2005; Liu et al. 2007). SAPALDIA focuses on health effects from long-term exposure to air pollution within a Swiss cohort of over 8,000 participants mainly living near eight study areas (Geneva, Basel, Lugano, Aarau, Wald, Payerne, Davos, Montana), where continuous air pollution measurements were done. Some of the participants moved house between the two health survey years (1991 and 2002) and are therefore dispersed over Switzerland. For these participants the output of a high resolution air quality model is needed to obtain their air pollution exposure. The high resolution domain of Switzerland is nested into a coarser European one. This study presents a sensitivity evaluation for July 2005 on the effects of different chemical and meteorological boundary and initial conditions on the model output of the coarser European domain. Therefore, this study presents a preparatory stage for the final overall aim. July 2005 was simulated because of the absence of GFS data for the years 1991 and 2002.

2 Method and data

2.1 Model description and setup

The Weather Research and Forecasting (WRF version 3.2.1) model with the research core (ARW) was used for this study (Skamarock and Klemp 2008). The European domain covers an area from northern Africa to southern Finland with a horizontal resolution of 50 km. This leads to a domain with 60 × 60 grid points for 27 vertical sigma layers. The microphysics were calculated with the Eta Ferrier scheme (Rogers et al. 2001). Other used physics options were the Bets-Miller-Janjic cumulus scheme (Janjic 1994, 2000), the Eta surface layer scheme (Janjic 1996, 2001), the Noah land-surface model (Chen and Dudhia 2001) with the USGS landuse dataset, the Mellor-Yamada-Janjic planetary boundary layer scheme (Janjic 1990, 1996, 2001), the RRTM (Mlawer et al. 1997) and Dudhia scheme (1989) for the longwave and shortwave radiation, respectively. Further informations on these options can be found in Skamarock et al. (2008).

The air pollutants were simulated with the chemistry extension (WRF-Chem version 3.2.1) (Grell et al. 2005). This model is widely used in America and was already used in Europe by Schürmann et al. (2009), SanJosé et al. (2008) and Tuccella et al. (2012). Recently, the model is more and more used by European research groups which can be seen in contributions to international conferences (e.g. EGU2011). WRF-Chem is an online coupled model with several choices of chemical mechanisms and aerosol modules. A small pre-study was carried out to choose the various options (e.g. chemical mechanism, physic parameters, dynamic parameters and other chemical options) of the WRF-Chem framework. For this study the best results were achieved with the Carbon bond mechanism version Z (CBMZ) and the corresponding Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) using four sectional aerosol bins (Zaveri and Peters 1999; Zaveri et al. 2008; Fast et al. 2006). Slightly better results were achieved with eight sectional bins but considering the additional computation time and the only little gain we decided to use only four bins. The same time steps were applied for the meteorological and the chemical calculations. The lumped CBMZ scheme consists of over 65 prognostic species and over 160 chemical reactions. The MOSAIC scheme implements primary aerosols (sea salt, soil dust, black carbon and organic carbon) as well as secondary aerosols formed by various salts, inorganic gas-phase chemistry, coagulation and homogeneous nucleation. The Fast-J photolysis (Wild et al. 2000) option was applied along with a dry deposition option. The setup of this domain did not implement direct and indirect radiative feedback, wet scavenging or the formation of secondary organic aerosols, because these sub-modules were not compatible with the chosen physics options and aerosol module, respectively.

2.2 Emissions

Gap-filled emissions from the European Monitoring and Evaluation Programme (EMEP) were used (Vestreng and Klein 2002; EMEP 2010) as anthropogenic input for July 2005. They are available with a horizontal resolution of 50 km and yearly average values of several main air pollutants, heavy metals, persistent organic pollutants and particulate matter for 11 SNAP sectors (Selected Nomenclature for reporting of Air Pollutants). For this study only the main pollutants (CO, NH_3, NMVOC, NO_x, SO_x) and the particulate matter ($PM_{2.5}$, PM_{coarse}) were used. The data were temporally disaggregated into hourly emissions according to the SNAP sectors, the country codes and the

time factors from the GENEMIS project (Friedrich and Reis 2004; Tuccella et al. 2012). Other converting steps were the vertical disaggregation into the lowest six model layers, the spatial conversion with the inverse next neighbor method and the chemical species disaggregation from the EMEP system into the used CBMZ system. A more detailed description of these processing steps can be found in Ritter (2009).

Biogenic emissions are built up on runtime using the model of emissions of gases and aerosols from nature (MEGAN version 2.0.4) (Guenther et al. 2006; Sakulyanontvittaya et al. 2008). The online implementation of the MEGAN emissions are slightly simplified in comparison to the standalone version. Only for isoprene emissions a separate map of emission factors is used. All other ones are assigned an emission factor based on the plant functional type. Also no soil moisture factor and no production and loss of emissions within the plant canopy are applied as no explicit canopy model is used.

2.3 Chemical boundary conditions

The effect of two different chemical boundary conditions on the European domain was evaluated against each other. The first one is hardcoded in the WRF-Chem model. The values are based on an idealized, northern hemispheric, mid-latitude, clean environmental, vertical profile from the NOAA Aeronomy Lab Regional Oxidant Model (NALROM) (McKeen et al. 1991; Liu et al. 1996; Peckham et al. 2010). This profile is declared globally so that lateral boundary conditions can be derived. Chemical initial conditions are not needed when a spin-up calculation of 5 days precedes. Consequent runs can be initialized by their already calculated chemical fields.

On the other hand, monthly mean values (1997–2001) of the global LMDZ-INCA model were implemented (Hauglustaine et al. 2004; Szopa et al. 2009) as chemical boundary conditions. This model is a coupled chemistry and aerosol model with a horizontal resolution of $3.75° \times 2.5°$. The weighted mean of the surrounding eight grid points (3D) is calculated for every boundary grid point of the European domain. This procedure was applied for eight different chemical species (O_3, NO, NO_2, HNO_3, PAN, H_2O_2, CO, HCHO). For all other chemical species the hardcoded values were taken as boundary conditions.

2.4 Meteorological input parameters

Two different meteorological input parameters were used for this study and their effect on the domain has been compared. The meteorological input parameters were newly initialized every five simulated days with an additional spin-off time of 12 h. This leads to a 5.5-day forecast, where the first 12 h where skipped to minimize the differences between the small scale features within the domain and the large scale features from the meteorological input datasets. The first ones are datasets of the Global Forecast System (GFS) that is available from NOAA. The horizontal resolution of the gridded dataset is 1.0° (original spectral grid: T382L64) for July 2005 and is available in 3 h steps. Only the initial and first forecast step were used to stay as close as possible to observations. Data is available online from February 2005 onwards.

Because the overall aim is to simulate the years 1991 and 2002 other meteorological input datasets namely Reanalysis data from NCEP were used. These datasets are continually gridded and incorporate observations and numerical weather prediction model output and date back to 1948. However, revised Reanalysis data (Reanalysis II) are available from 1979 onwards with a horizontal resolution of 2.5° (original spectral grid: T62L28) for 6 h timesteps (Kanamitsu et al. 2002). For this study the revised Reanalysis II data were compared to the GFS data to evaluate how and to what extent the two different datasets influence modeled air pollutants within WRF-Chem.

2.5 European measurements

The validation of the model results was done with data from the EMEP Measurement Network for O_3, SO_2, NO_2, PM_{10} and $PM_{2.5}$ (EMEP 2010). All EMEP stations are ground-based and classified as rural and additionally only stations with a fixed minimum distance to the boundaries were considered to eliminate artifacts from boundary conditions. Overall around 100 O_3, 40 SO_2, 30 NO_2 and PM_{10} and 17 $PM_{2.5}$ stations could be used for the verification (see Fig. 1). $PM_{2.5}$ measurements were done at the same location as some PM_{10} measurements and therefore not shown in Fig. 1. The validation for SO_2 was not taken into account, because SO_2 concentrations for the European domain are very low and of little interest for the overall study.

2.6 Statistical indicators

There are a number of statistical parameters that can be used to validate model output. This study focused on the Pearson correlation coefficient, the mean bias, the root mean square error and the mean absolute gross error. These parameters were often used and suggested in various scientific literature (deMeij et al. 2009; Seinfeld and Pandis 2006; Jacobson 2005).

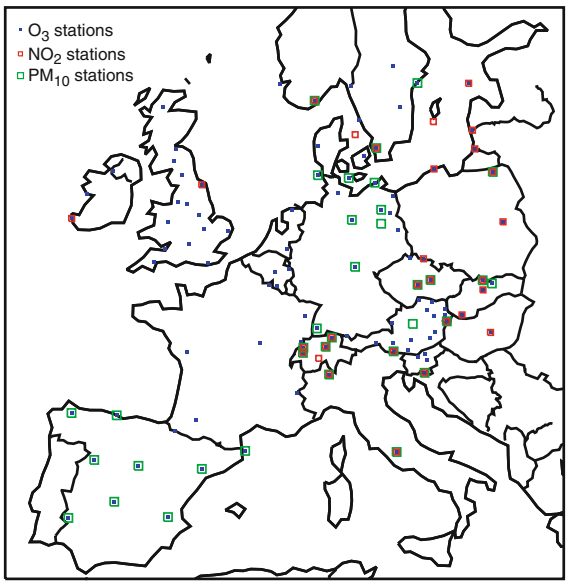

Fig. 1 EMEP measurement stations (O_3, NO_2 and PM_{10}) with valid data for the modeled time period (July 2005)

3 Results

3.1 Chemical boundary conditions

We compared the influence of different chemical boundary conditions (LMDZ-INCA vs. idealized profiles) on WRF-Chem (meteorological initialization with GFS). Fig. 2 shows the mean difference (LMDZ-INCA minus NALROM) of ground-level ozone for these two runs for July 2005 and the corresponding scatterplot with the LMDZ-INCA-based runs on the *x* axis and the NALROM-based runs on the *y* axis. Almost over the whole domain we see a positive mean difference up to 25 µg/m^3. Only over the north-western part of the domain (Atlantic Ocean) we notice negative differences of ozone concentration up to 12 µg/m^3. A big distinction between the two chemical boundary conditions is the availability of spatial data (LMDZ-INCA) versus only a globally applied, vertical profile (NALROM). Figure 3 shows the monthly mean July concentrations (1997–2001) for ground-based ozone from the LMDZ-INCA model for the European domain. The standardized profile (NALROM) has a concentration of 0.03 ppmv (around 60 µg/m^3) for ground-level ozone at standard atmospheric pressure. With a special focus on the borders of this figure (boundary conditions) we observe significant higher ozone concentrations over land-based pixels compared to the 60 µg/m^3 and in contrast lower concentrations over the Atlantic Ocean. The distribution of ozone concentrations at the boundaries of Fig. 3 explains the differences of the model output initialized by the standardized profile and the LMDZ-INCA model. A correlation of 0.90 between the runs initialized by LMDZ-INCA and NALROM is achieved. If we subtract 300 km of each boundary (equals 10% on each side) the correlation slightly goes up to 0.93 (rmse = 2.00, mage = 0.33, bias = 0.28). Table 1 indicates the statistical values of the two runs (NALROM + GFS and LMDZ-INCA + GFS) compared to ozone EMEP measurements. Chemical boundaries derived from the LMDZ-INCA model perform better than the standardized profiles in terms of the ozone results of WRF-Chem. We can observe this fact in the Pearson correlation coefficient for the hourly data comparison (0.63 vs. 0.60), the daily mean values (0.67 vs. 0.62) and the daily maxima values (0.72 vs. 0.64) for the LMDZ-INCA initialized run and the NALROM initialized run, respectively.

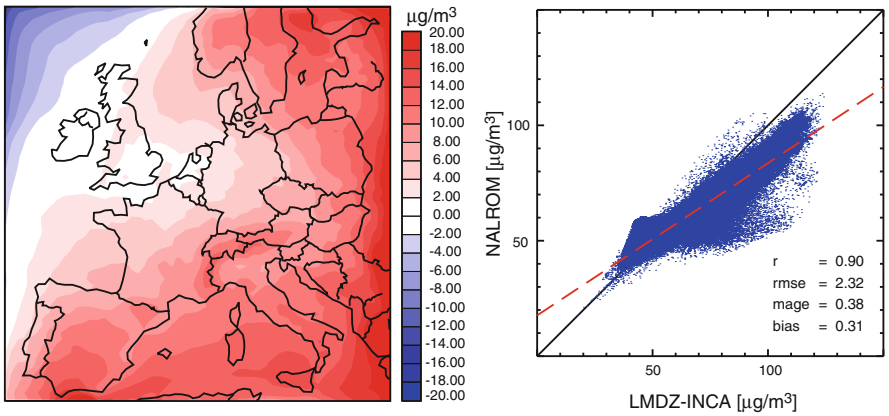

Fig. 2 Map (*left*) and scatterplot (*right*) of the mean difference of ground-level ozone (O_3) simulated for July 2005 with WRF-Chem driven by monthly mean values from the LMDZ-INCA model minus WRF-Chem driven by standardized profiles obtained from the NALROM model as chemical boundary conditions

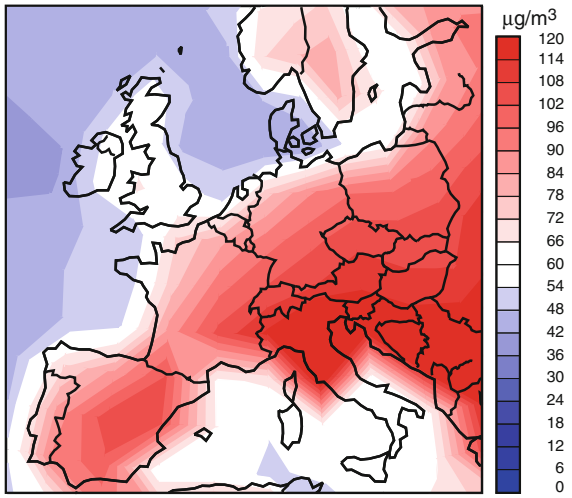

Fig. 3 Monthly mean values for July (1997–2001) of ground-level ozone (O_3) from the LMDZ-INCA model

Other than the standardized profile the LMDZ-INCA model represents the tropopause with very high ozone concentrations in the stratosphere. Therefore, huge differences in ozone concentrations can be found in upper levels between these two runs. Already from 800 hPa on only positive mean difference can be found in favor of the model output initialized by LMDZ-INCA. As the overall aim of this study is to compare air pollutants on health effects, we will furthermore focus on ground-based results.

The same comparison of the effect of the two different chemical boundary conditions on WRF-Chem was made for NO_2 (Fig. 4), PM_{10} and $PM_{2.5}$. Boundary pixels for NO_2 at ground level derived from the LMDZ-INCA model have a range of approx. 0.003–3.5 µg/m^3 whereas the standardized NALROM profile has a value of approx. 0.03 µg/m^3. In the middle and upper troposphere the concentrations are roughly the same. Almost no influence on the mean difference of the two modeled runs can be seen in Fig. 4. Only over the southern part of the North Sea and at the boundaries some regions with positive differences can be found. These differences never exceed more than 0.8 µg/m^3 and the Pearson correlation coefficient tends to 1.00. As PM_{10} and $PM_{2.5}$ simulations were absent in the LMDZ-INCA model, the same boundary values were applied as the ones from the standardized profile. Pearson correlation coefficient tends to 1.00 for PM_{10} as well as for $PM_{2.5}$. Therefore, the figures are not shown in this paper. However, there are marginal differences of up to 1 µg/m^3 for PM_{10}. Nitric acid (HNO_3) is available by the LMDZ-INCA model which can react as precursor for particulate matter. How far this contributes to the differences could not be evaluated in this study.

3.2 Meteorological initial and boundary conditions

The influence of different meteorological initial and boundary conditions (GFS and Reanalysis II) on ozone concentrations of WRF-Chem (LMDZ-INCA as chemical boundary conditions) are shown in Fig. 5. These options were tested due to the absence of GFS data for the intended years of the overall study (1991 and 2002). The map shows the mean differences in µg/m^3 of ozone for July 2005 (GFS minus Reanalysis II). The values rarely exceed a difference of more than 5 µg/m^3 in the center of the domain. These small differences already occurred at the first modeled time step and are transported along with the wind at roughly the same directions for both different runs. Summarized over the entire month land-based pixels show rather a positive difference and water pixels rather a negative one. The scatterplot on the right side of Fig. 5 represents WRF-Chem initialized by the GFS (x axis) and Reanalysis II (y axis) data. Overall a correlation of 0.96 is achieved, which leads to the assumption that we can use the Reanalysis II data for the overall study without any concerns regarding ozone. Despite the larger discrepancies near the eastern boundaries and especially near the south of Spain the statistical values remain exactly the same if we subtract 300 km from each side of the domain for the analysis. However, the difference near the south of Spain can be explained by a difference in near-surface

Table 1 Comparison of the statistical values for ozone (July 2005) at the EMEP measurements and ozone computed with WRF-Chem initialized by different chemical and meteorological boundary and initial conditions

Frequency	IC/BC	Bias	Mage	rmse	r
O_3 hourly values	NALROM + GFS	−10.11	20.65	26.84	0.60
	LMDZ-INCA + GFS	−3.15	18.53	24.13	0.63
	LMDZ-INCA + R2	−3.81	18.82	24.52	0.62
O_3 daily means	NALROM + GFS	−10.19	16.15	21.57	0.62
	LMDZ-INCA + GFS	−3.24	13.83	17.99	0.67
	LMDZ-INCA + R2	−3.90	14.11	18.41	0.66
O_3 daily maxima	NALROM + GFS	−19.19	23.23	30.09	0.64
	LMDZ-INCA + GFS	−11.33	18.04	23.87	0.72
	LMDZ-INCA + R2	−12.34	18.81	24.74	0.70

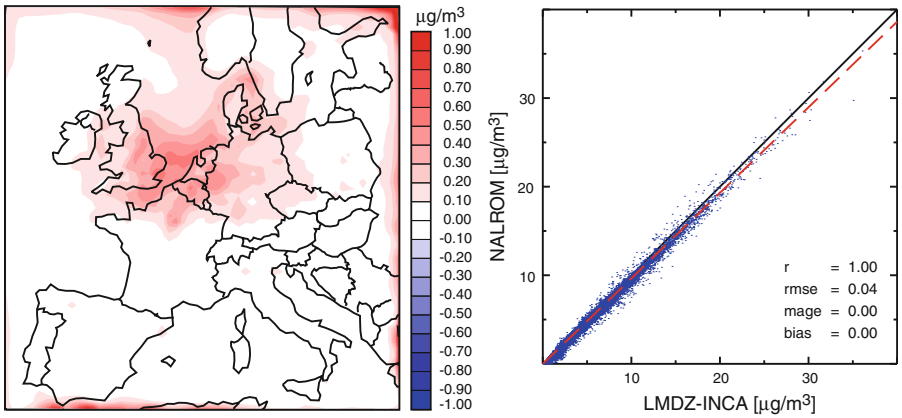

Fig. 4 *Map* (*left*) and *scatterplot* (*right*) of the mean difference of ground-level nitrogen dioxide (NO_2) simulated for July 2005 with WRF-Chem driven by monthly mean values from the LMDZ-INCA model minus WRF-Chem driven by standardized profiles obtained from the NALROM model as chemical boundary conditions

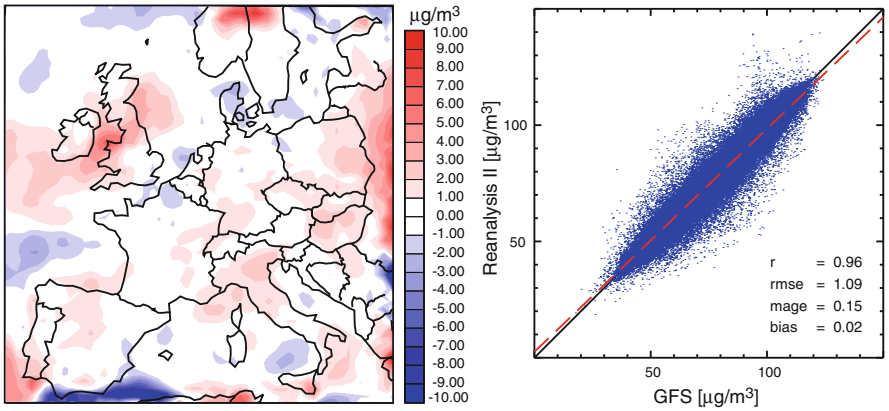

Fig. 5 *Map* (*left*) and *scatterplot* (*right*) of the mean difference of ground-level ozone (O_3) simulated for July 2005 with WRF-Chem initialized by GFS data minus WRF-Chem initialized by Reanalysis II meteorological data

temperature of up to 5 K in the same region when the model was initialized by Reanalysis II instead of GFS. All other differences can neither be explained by differences in temperature nor solar radiation. Compared to the EMEP measurements the model output initialized by the GFS model performed slightly better (see Table 1) for ozone. The Pearson correlation coefficient for the hourly data, daily mean and daily maxima of O_3 were 0.63, 0.67 and 0.72 for the GFS model output and 0.62, 0.66, 0.70 for the Reanalysis II model output.

The two meteorological input parameters were also tested for NO_2, PM_{10} and $PM_{2.5}$, whereas the latter one is not presented in this paper due to its similarity to PM_{10}. NO_2 differences of the two runs initialized by GFS and Reanalysis II occur near coastal regions, over land, at the southern boundaries and particularly between the area of southern England and the Netherlands (see Fig. 6). Likewise for O_3, the differences in the center of the domain are mainly caused by the meteorological initial parameters and the ones at the boundaries mainly by the boundary conditions. The coastal and boundary differences can be explained with the higher resolution of GFS and its better representation of the meteorological (especially over the coastal and boundary regions). The mean differences (positive and negative) of more than 1 µg/m^3 NO_2 in the area of southern England and the Netherlands over the whole July 2005 were due to several anticyclone systems that were not properly captured with either the GFS or the Reanalysis II dataset. Nevertheless, a Pearson correlation coefficient of 0.96 between these two runs could be achieved. For PM_{10} only a Pearson correlation coefficient of 0.90 ($PM_{2.5}$: 0.89) has been obtained. The points of the

scatterplot (Fig. 7) disperse a lot more and on the map are larger regions with higher differences between the two runs initialized by GFS and Reanalysis II. There is no clear pattern as it can be seen for O_3 or NO_2 and the differences cannot be explained by looking at the distribution or the differences of near-surface temperature and solar radiation. The differences occur from the first timestep on but increase during the run, so that the boundary conditions seem to have a bigger influence as for the other two pollutants.

3.3 Evaluation with measurements

In Fig. 8a–d, hourly mean values of all available EMEP measurement stations and the mean of their corresponding grid points can be seen for July 2005. The results were obtained using the LMDZ-INCA model as chemical boundary conditions and the GFS model as meteorological initial and boundary conditions. For ozone good correlation exists (part A), also due to its dependency on meteorological parameters (e.g. temperature and solar radiation). Part B shows NO_2 with the additional line being the daily average of the hourly model output data for a better comparison as NO_2 stations were only available as daily values. WRF-Chem simulates too low concentrations over the entire month but is still within a satisfying range if we take into account that the horizontal resolution is 50 km with EMEP stations classified as rural. Trends can more or less be captured by the model. Part C and D show particulate matter (PM_{10} and $PM_{2.5}$) also with an additional line as modeled daily mean values. Both modeled pollutants have a similar trend as well as the corresponding observations.

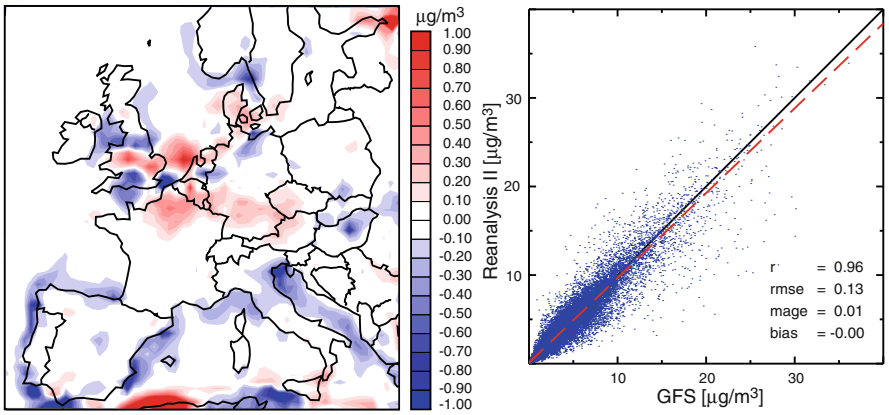

Fig. 6 *Map* (*left*) and *scatterplot* (*right*) of the mean difference of ground-level nitrogen dioxide (NO_2) simulated for July 2005 with WRF-Chem initialized by GFS data minus WRF-Chem initialized by Reanalysis II meteorological data

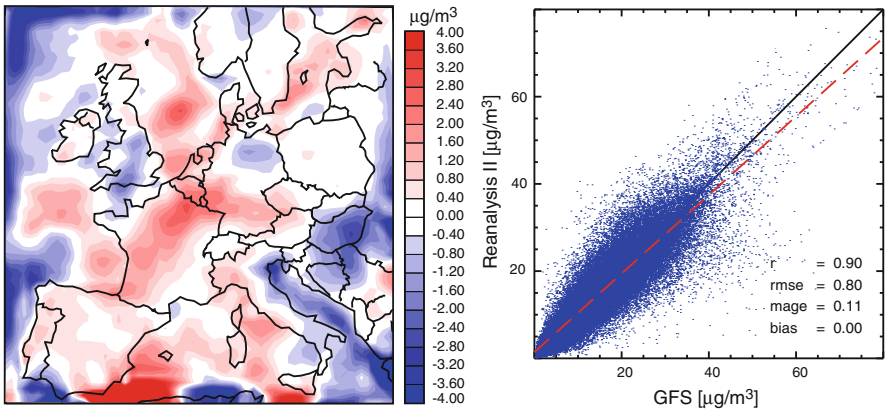

Fig. 7 *Map* (*left*) and *scatterplot* (*right*) of the mean difference of ground-level particulate matter (PM_{10}) simulated for July 2005 with WRF-Chem initialized by GFS data minus WRF-Chem initialized by Reanalysis II meteorological data

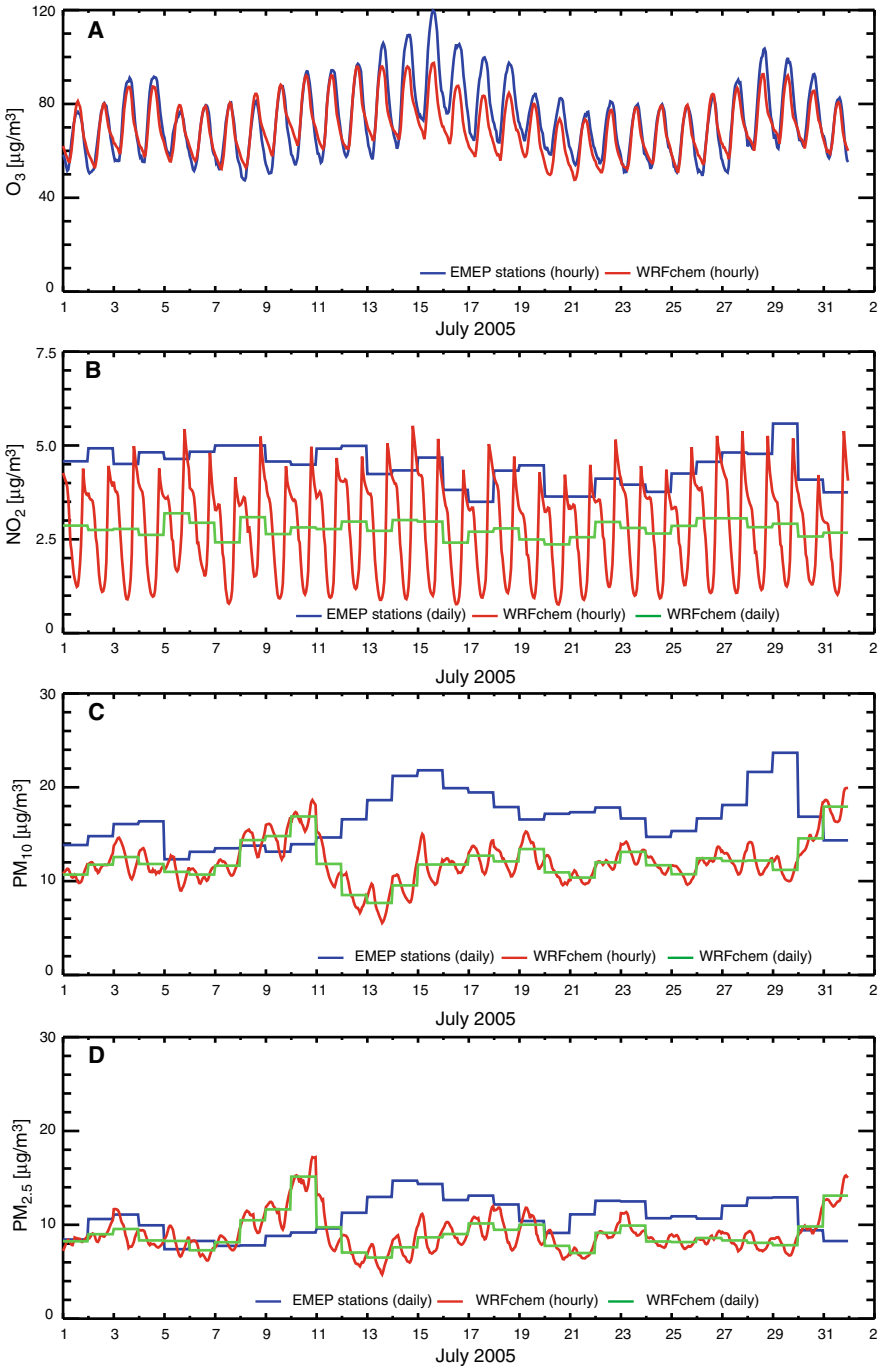

Fig. 8 Mean of EMEP measurements and mean of their corresponding model grid points of WRF-Chem (hourly and daily values) for July 2005. **a** O_3, **b** NO_2, **c** PM_{10} and **d** $PM_{2.5}$

Nevertheless, the trends have a smaller overall amplitude for modeled and measured data for $PM_{2.5}$ compared to PM_{10}. Both pollutants have periods of similar and controversial trends between the model output and the observations. We could not find any patterns for the near-surface temperature or solar radiation that could explain these differences between the model and the measurements. The differences are sometimes more than 25 µg/m^3 between a single station (especially some stations in Spain) and the corresponding modeled grid point. The meteorological conditions during the modeled month (July 2005) caused severe droughts in Spain, Portugal and southern France and

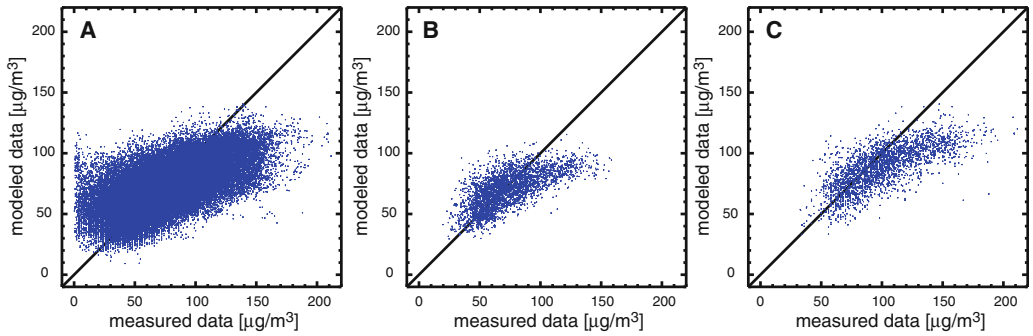

Fig. 9 Scatterplots for O_3 hourly values (**a**), O_3 daily mean (**b**) and O_3 daily maxima (**c**) for July 2005. Illustrated are EMEP measurements(x axis) against their corresponding grid points of the WRF-Chem output (y axis)

wildfires in Spain. How far these wildfires contributed to the PM_{10} concentrations or the differences between the model and the EMEP stations could not be shown in this study.

For ozone further analysis was done as hourly observations were available. Scatterplots for hourly data, daily mean and daily maxima are illustrated in Fig. 9a–c. The measured data forms the x axis and the modeled data the y axis (LMDZ-INCA and GFS as boundary and initial condition). Corresponding statistical parameters are listed in Table 1. Every available hourly data of all EMEP measurements were statistically analysed (over 100). The Pearson correlation coefficient increases from 0.63 to 0.67 and 0.72 for hourly data, daily means and daily maxima, respectively.

4 Discussion

The sensitivity test for the different chemical boundary conditions showed that there are differences between the two runs concerning ozone. The differences of up to 60 µg/m^3 between the different boundary conditions are decreased after the calculations. Only a small region over the Atlantic Ocean has a negative difference between the two runs but also these negative differences disappear from 800 hPa on upwards. The use of spatially varying chemical boundary conditions (e.g. LMDZ-INCA) with its representation of the tropopause increased the performance of WRF-Chem as the model fitted better with EMEP measurements. Nevertheless, the influence of different chemical boundary conditions on other pollutants (e.g. NO_2, PM_{10} and $PM_{2.5}$) seems negligible as Pearson correlation coefficients of 1.00 were achieved between the two different initialized runs. For PM_{10} this is not surprising as data is not available in the LMDZ-INCA model and the same values from the standardized profile were taken. Also for NO_2 the differences between the two runs are minor. The standardized profile has a concentration around 0.03 µg/m^3 for ground-level NO_2 whereas the LMDZ-INCA values vary between 0.003 and 3.5 µg/m^3 at the boundaries of the domain. The chemical boundary conditions do not have a strong impact on NO_2 concentrations as the anthropogenic emissions dominate. According to Szopa et al. (2009) better results compared to measurements could be possible with more refined, daily chemical boundary conditions, especially for regions near the boundaries.

The comparison of the use of different meteorological initial and boundary conditions revealed minor mean differences for ozone and NO_2. All the graphs shown in this paper exclude a spin-off time from June 26th to 30th. However, already these spin-off days showed almost the same difference as the mean concentrations of July. Therefore, the differences for O_3 and NO_2 are mostly due to the initial field and not due to the meteorological boundary parameters. The minor discrepancies between the runs are probably caused by the different horizontal resolutions or anticyclone systems that were not correctly represented within one of the two meteorological datasets. For these two pollutants (O_3 and NO_2) the decrease in accuracy of the model is small and Reanalysis II data can be used for the European domain without any concern. However, the Pearson correlation coefficient for particulate matter was only 0.90 (between the two different runs) and differences up to 5 µg/m^3 exist at the boundary regions of the domain. Therefore, the use of different meteorological input parameters leads to a change in PM_{10} concentration. The statistical parameters for O_3, NO_2 and PM_{10} did not change with the exclusion of the boundaries of the domain in the analysis. Overall, GFS data should be preferred over Reanalysis II datasets when possible. On the one hand, the correlations of air pollutants to EMEP measurement stations were slightly better when WRF-Chem was initialized by GFS (see Table 1 for O_3) and on the other hand, the meteorological parameters (e.g. temperature) also achieved better correlations compared to weather stations. Commonly, more often updated meteorological boundary

conditions (3 h for GFS instead of 6 h for Reanalysis II) are more suitable for limited domains.

Regarding the comparison of the results to measurements, the model performs well for ozone. The diurnal cycle is well represented and trends are captured by WRF-Chem. For NO_2 some of the trends can be represented by the model (e.g. July 20th–26th in Fig. 8b). On the other hand, during the entire month, the concentrations were underestimated with absolute difference between 1 and 3 $\mu g/m^3$. Due to its dependencies on a variety of parameters, PM_{10} seems to be difficult to model over a longer period. The first days are in good agreement with measured data (e.g. July 1st–11th in Fig. 8c), but afterwards contrasting trends are observed even though the values of the modeled PM_{10} are within an acceptable range. We further looked at meteorological parameters to find an explanation for the contrasting trends but neither temperature, solar radiation nor wind speed could explain the trends. When we looked at individual stations and their corresponding modeled grid point we see that not all stations have these controversial trends. On the other hand, big differences between model and observation can be found for some stations in Spain. How far for example wildfires contribute to these differences, cannot be shown in this study. The same facts can be observed for $PM_{2.5}$ but with lower absolute differences between the model and the measurements. Trends seem to be identical for particles smaller than 10 and 2.5 µm. According to Seinfeld and Pandis (2006) and Hallquist et al. (2009), the implementation of an additional secondary organic aerosol module (e.g. Schell et al. 2001) would have significant influence on concentrations of particulate matter. The constant factors for splitting $PM_{2.5}$ and PM_{coarse} emissions into aerosol components (elemental carbon, organic aerosol, sulfate, nitrate and undefined mass) and different sizes (Aitken and accumulation mode for $PM_{2.5}$ and afterwards the 4 different bins) also have an impact on particle concentrations. Furthermore, the number of EMEP measurement stations (approx. 30 for PM_{10} and NO_2) and their location could have an influence on the comparison. Even though all EMEP stations are classified as rural, it is not always self-evident that one station can correctly represent an area of 50 km × 50 km. This issue is exacerbated when the altitude difference of the station and the corresponding grid point increases. On the other hand, an accumulation of stations within a small region leads to an overweighing of some region (e.g. Switzerland, Austria, United Kingdom).

A good overview of the performance of other air quality models over Europe and their statistical values can be found in Pay et al. (2010) and more detailed model performance statistics for some models in van Loon et al. (2004, 2007). The range of the correlation factors for most of the studies in the aforementioned papers is within 0.55–0.8 for ozone daily averages and 0.69–0.84 for daily peak values. We have to take into account that this paper only presented the output of July 2005 and not an entire year. The data in van Loon et al. (2007) are, in addition to being presented as annual statistical values, also summarized into seasonal data. With regard to the differences of seasonal and monthly data, we tried to compare the Pearson correlation coefficient of this paper with the seasonal ones from van Loon et al. (2007). For daily average WRF-Chem (0.67) performed better than all the other mentioned models (0.35–0.64) and only slightly worse than their ensemble (0.68). Daily maxima values for the models in van Loon et al. (2007) are between 0.51 and 0.77 and the ensemble is 0.78. Only the CHIMERE model and the ensemble performed slightly better than WRF-Chem in this paper. We have to mention that the models used in van Loon et al. (2007) were compared to a reduced set of stations in order to prevent overweighing of some regions with a large number of stations and only stations below an altitude of 1,000 m were considered. We also tested the performance of WRF-Chem compared to stations only below a certain altitude but there were no significant changes so that we preferred to keep as much stations as possible for the comparison.

5 Conclusions

This paper presented the influence of different chemical and meteorological boundary and initial conditions on air pollutants and their comparison to EMEP measurements. Chemical boundary conditions have mainly an influence on ozone and meteorological initial and boundary conditions have the biggest impact on particulate matter. The Pearson correlation coefficients range from 0.90 to 1.00. The monthly mean values driven by the LMDZ-INCA model performed better than the time invariant standardized profile compared to measurements. The influence of meteorological initial and boundary conditions on ozone concentrations was not as significant as the chemical boundary conditions but still noticeable. For NO_2 the differences between the two runs were rather small. A noticeable impact on the two runs with different meteorological input parameters is found for particulate matter. Nevertheless, summarized over the entire European domain, the NCEP Reanalysis II data produced satisfying results even though the GFS model with the higher resolution leads to slightly better results. Whenever possible GFS data should be preferred as meteorological input parameters, especially for regional domains. For the years before 2004, Reanalysis II data are a good alternative to initialize WRF-Chem. The European simulations show good results for observed air pollutants, with ozone being

the most and PM_{10} being the least satisfying. In comparison to statistical values of other models used over Europe for ozone, WRF-Chem shows encouraging results. The flexibility of WRF-Chem with its modular design allows the user to implement different chemical options and to test them easily. Improvements in the model output could probably be achieved with more detailed daily chemical boundary conditions, more refined chemical species conversion, a better horizontal and vertical resolution and/or more detailed anthropogenic emissions. If more rural measurement stations for other air pollutants in addition to ozone existed (in particular, hourly measurements), a better validation could be made. To conclude, WRF-Chem performed best with spatially resolved chemical boundary conditions and high resolution meteorological input parameters. The model performed satisfyingly for observed pollutants and is within the top air quality models.

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3.2. Swiss domain

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Air pollution modeling over very complex terrain. A Swiss two year evaluation of WRF-Chem.

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Abstract

The Weather Research and Forecasting (WRF) model with its online chemistry extension (WRF-Chem) has been implemented over a Swiss domain for the years 2002 and 1991. The very complex terrain requires a high horizontal resolution (2x2 km^2), which is achieved by nesting the Swiss domain into a coarser European one. A multi-linear regression post-processing serves to eliminate systematic bias. The temporal and spatial distribution of O_3, NO_2 and PM_{10} as well as temperature and solar radiation are evaluated against ground-based measurements. The model performs well for the meteorological parameters with Pearson correlation coefficients of 0.92 for temperature and 0.88-0.89 for solar radiation. Seasonal variations of air pollutants are represented correctly. However, short-term peaks of several days are not captured by the model. Averaged O_3 has satisfying results for daily mean values and daily maximum values (0.67-0.75) whereas averaged NO_2 and PM_{10} have best scores for yearly averaged values (0.71-0.80). The spatial distribution reveals the importance of PM_{10} advection from the Po valley to the canton of Ticino. For all pollutants the absolute station errors are small. Larger errors occur along heavy traffic roads, in street canyons or on mountains. We also compare yearly modeled results against a dedicated Swiss dispersion model for NO_2 and PM_{10}. Both models have similar results, but WRF-Chem is capable of computing the temporal evolution of three-dimensional data for a variety of air pollutants and meteorological parameters. Overall, WRF-Chem produces encouraging results over very complex terrain with the application of post-processing algorithms.

Keywords: air pollution modeling, air quality, WRF-Chem, Switzerland, PolluMap

1. Introduction

Air pollutants are part of a very complex system, which affects human health, climate change and the physics and chemistry of the atmosphere. Especially the first point is gaining more and more attention as longterm cohort studies are showing clear evidence of negative health effects (Dockery et al., 1993; Ackermann-Liebrich et al., 1997; Künzli et al., 2000; Pope et al., 2002; Ackermann-Liebrich et al., 2005; Filleul et al., 2005; Gehring et al., 2006; Vineis et al., 2006; HEI, 2010). To assess individual exposure of health study participants there exist different methods. Most studies generally use a central-site measurements to represent cohort exposures. More recent studies are using dedicated dispersion models, land-use regression models or chemical transport models (CTM), whereas the latter is the most complex approach. CTM's are often separated into offline and online models. The offline ones use a separated numerical weather prediction (NWP) model with a dedicated CTM model, whereas the online approach is a combination of these two models together. Further information on the definition and also a listing of different models can be found in Kukkonen et al. (2012); Pay et al. (2010); van Loon et al. (2007, 2004); Vautard et al. (2009). In this study we applied the state-of-the-art Weather Research and Forecasting (WRF) model (Skamarock et al., 2008;

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Skamarock and Klemp, 2008) with its chemistry extension (WRF-Chem) to simulate air pollutants (Grell et al., 2005). Together they form an online approach with a variety of different physics, dynamics and chemistry options. Recently this model was applied over Europe by San José et al. (2008); Schürmann et al. (2009); Tuccella et al. (2012); Webley et al. (2012).

The overall aim of this study is to characterize the spatio-temporal distribution of multiple air pollutants for the Swiss cohort study on air pollution and lung diseases in adults (SAPALDIA) for the years 1991 and 2002 (Ackermann-Liebrich et al., 1997, 2005; Bayer-Oglesby et al., 2006; Liu et al., 2007). SAPALDIA focuses on health effects from long-term air pollution exposure within a Swiss cohort of over 8000 participants. Liu et al. (2007, 2012) show the techniques thus far used for assessing the individual exposure of the participants. Swiss wide estimations were modeled annually by a dedicated dispersion model (PolluMap) with a horizontal resolution of 200 m (SAEFL, 2003a, 2004b). This model approach is limited to NO_2, PM_{10} and $PM_{2.5}$. WRF-Chem was applied to model more air pollutants at a higher temporal resolution using more refined meteorological input data. WRF-Chem was mainly chosen for its online application. To our knowledge, the use of such a model to estimate air pollution exposure of a longitudinal cohort will be unique. There exists at least two other chemical transport models for Switzerland using offline approaches. One is a model based on the offline model CHIMERE with MM5 for a horizontal resolution of 15 km operated by GAIASENS Technologies Sarl (Couach et al., 2004; Kirchner et al., 2001). Another is operated by the Paul-Scherrer-Institue which uses an offline model (CAMx) coupled with WRF (Andreani-Aksoyoglu et al., 2008; Keller et al., 2008; Aksoyoglu et al., 2011).

In this paper, we first describe the applied models, their setup and the used emissions in section 2. Then we explain the application of a post-processing approach in section 3. Following a short validation of the modeled temperature and solar radiation, we validate O_3, NO_2 and PM_{10} for the years 1991 and 2002 (see Sec 4). The graphs mainly focus on the year 2002. Finally, we compare our WRF-Chem results with those of the existing PolluMap model.

2. Method and Data

2.1. Model description

The Weather Research and Forecasting (WRF) model version 3.2.1 with its research core (ARW) is used for simulating the meteorology. The model is a mesoscale, fully compressible, Eulerian non-hydrostatic model with several options for physical parameterization (Skamarock and Klemp, 2008; Skamarock et al., 2008). The chosen options are listed in Tab. 1. Microphysical processes are treated by the Eta grid-scale cloud and precipitation scheme (Eta Ferrier), which predicts changes in water vapor and condensate in cloud water, rain, cloud ice and precipitation ice (Rogers et al., 2001). The cumulus parameterization for sub-grid-scale effects of clouds is realized by the Betts-Miller-Janjic scheme (Janjic, 1994, 2000). The surface layer scheme (Eta) is based on the Monin-Obukhov similarity theory and described in Janjic (1996, 2001). The Noah Land-Surface scheme (Chen and Dudhia, 2001) along with USGS landuse dataset are used and the Mellor-Yamada-Janjic (Janjic, 1990, 1996, 2001) scheme is applied for the representation of the planetary boundary layer. Radiation is simulated by the Rapid Radiative Transfer Model (RRTM) for longwave (Mlawer et al., 1997) and the Dudhia scheme for shortwave (Dudhia, 1989) radiation, respectively.

Table 1: Physical parameterization of the Weather Research and Forecasting (WRF) model.

Process	WRF option
Microphysics	Eta Ferrier scheme
Cumulus parameterization	Bets-Miller-Janjic
Surface Layer	Eta similarity theory
Land-Surface model	Noah land-surface model
Landuse dataset	USGS
Planetary Boundary Layer	Mellor-Yamada-Janjic PBL
Longwave radiation	RRTM
Shortwave radiation	Dudhia scheme

For simulating air pollutants the online approach using the WRF chemistry extension (WRF-Chem) version 3.2.1 was applied (Grell et al., 2005; Fast et al., 2006). This approach has the advantage of using the same time steps, grid cells and transport scheme as WRF and is therefore fully consistent with the meteorological component. However, this also means that chemistry is calculated at every grid cell for every time step which strongly affects computing resources. WRF-Chem has several choices of chemical mechanism and aerosol modules. This study uses the Carbon bond mechanism version Z (CBM-Z) and the corresponding Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) using four sectional aerosol bins (Zaveri and Peters, 1999; Zaveri et al., 2008; Fast et al., 2006; Ritter, 2010). The CBM-Z model uses a lumped structure approach and contains over 65 prognostic species and over 160 chemical reactions. MOSAIC is used with a sectional four bins approach where each bin is assumed to be internally mixed. The module implements primary aerosols (sea salt, soil dust, black carbon and organic carbon) as well as secondary aerosols formed by various salts, inorganic gas-phase chemistry, coagulation and homogeneous nucleation. Photolysis frequencies are calculated with the Fast-J scheme (Wild et al., 2000) and dry deposition velocity is calculated using a approach developed by Wesely (1989). Several optional choices for WRF-Chem were not implemented in this study. Wet scavenging, direct and indirect radiative feedback was incompatible with some chosen physics options and the formation of secondary organic aerosols is only implemented from the WRF-Chem version 3.3 on for MOSAIC.

2.2. Model setup

A Swiss domain was simulated for the years 2002 and 1991. The horizontal resolution of the Swiss domain is approximately 2 km, which leads to 210x135 grid points (see Fig. 1). The vertical resolution is represented by 27 sigma coordinates from surface up to 50 hPa. The internal model time steps are 12 seconds and the model output is stored in hourly values. The simulations consist of modeled periods of 5 days with a meteorological spin-off time of an additional 12 hours. Initial chemical fields are directly taken from the preceding run. An extra 5 day period was calculated before the first run of the specific year to obtain initial chemical data. Meteorological initial and boundary conditions as well as chemical boundary conditions are derived through nesting the Swiss domain into a coarser European one. A one-way nesting approach was chosen for the convenience of being able to calculate the European domain prior to the Swiss domain.

The European domain covers an area from northern Africa to southern Finland with a horizontal resolution of 30 km and also 27 vertical sigma layers. The parent-grid ratio of 15 seems high compared to other studies. We therefore did an evaluation with only meteorological simulations with this ratio. The model performed stably and had satisfactory results. Chemical parameters are just passed by as a scalar field and do not interfere with the one-way nesting method. The European domain was calculated with the same versions of WRF and WRF-Chem and the same physical and chemical options. Internal time resolution for this coarser domain was every 3 minutes and the output was also stored in hourly values. Initial chemical conditions are, as in the Swiss domain, taken from preceding runs. Meteorological initial and boundary conditions are derived from Reanalysis II data from NCEP with a horizontal resolution of 2.5 degree (original spectral grid: T62L28) for six hour steps (Kanamitsu et al., 2002). Monthly mean values (1997-2001) of the global LMDZ-INCA model delivered the chemical boundary conditions (Hauglustaine et al., 2004; Szopa et al., 2009). The LMDZ-INCA model has a horizontal resolution of 3.75 degrees x 2.5 degrees. As the model could only be used for eight different chemical species (O_3, NO, NO_2, HNO_3, PAN, H_2O_2, CO, HCHO), all other needed species were taken from a hardcoded, vertical profile based on calculations of the NOAA Aeronomy Lab Regional Oxidant Model (NALROM) (McKeen et al., 1991; Liu et al., 1996; Peckham et al., 2010). Disaggregated emissions from the EMEP database (European Monitoring and Evaluation Programme) served as anthropogenic emissions (Vestreng and Klein, 2002; EEA, 2009) and biogenic emissions were built up on runtime from MEGAN version 2.0.4 (Guenther et al., 2006; Sakulyanontvittaya et al., 2008). Further details of the methodology can be found in Ritter (2009). A validation of the European domain to ground based measurements with specific focus on different meteorological initial and boundary conditions as well as different chemical boundary conditions can be found in Ritter et al. (2012).

The research core of the WRF model has stability problems when used over complex terrain (e.g. Switzerland) with a high horizontal resolution (e.g. 2 km). One option to resolve this problem is to increase the three-dimensional diffusion. However, we preferred to apply a smoothing to the high resolution topography data (USGS 30s) instead of modifying the whole meteorology through increased diffusion. We applied a hardcoded, smooth-desmooth algorithm from the WRF Preprocessing System (WPS). As we wanted to keep all information available in highly populated

regions of Switzerland (lower regions), we applied this smooth-desmooth process only for grid point with an altitude higher than 1000 m. For stable runs of WRF we had to iterate this algorithm four times. Fig. 1 shows the already smoothed topography. We can observe more spatial details in regions below 1000 m (e.g. northern part of the domain).

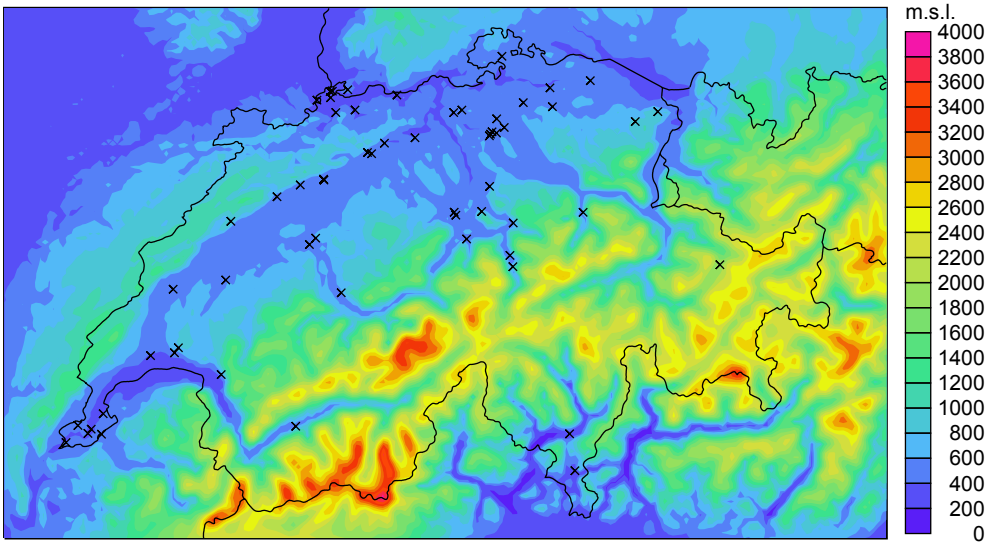

Figure 1: The Swiss domain with the location of all air pollution stations. Background is the altitude with smoothed values for grid points over 1000 m.

2.3. Swiss emissions

Anthropogenic emissions for the Swiss domain were obtained from SAEFL (2003a, 2004b) for PM_{10} and $PM_{2.5}$ as well as for NO_x. These emissions have a horizontal resolution of 200 m and are annual values for the year 2000. All emissions per pollutant have a source description with which the emissions could be split-up into 11 main SNAP sectors (Selected Nomenclature for reporting of Air Pollutants, version 1997) (Vestreng and Klein, 2002; EEA, 2009). Emissions for SO_2, NMVOC, CO and NH_3 were built-up with the same methodology as NO_x emissions from SAEFL (2004b). The same 200 m grid was constructed and the national total reference values were taken from SAEFL (1995, 2004a); FOEN (2008).

All annual emissions from the year 2000 are scaled to the Swiss national total amount as reported to the EMEP database (EEA, 2009) for each pollutant and SNAP sector of the intended years (1991 and 2002). Hourly emissions are achieved by a split-up according to the GENEMIS project with the help of the SNAP sectors (Friedrich and Reis, 2004). Further steps include a vertical disaggregation into the lowest 6 model layers, a spatial conversion with the inverse next neighbor method of 50 m tiles and a chemical species conversion from the afore mentioned pollutants to the used CBM-Z system. The disaggregations are basically the same as those used for the coarser European domain (Ritter, 2009; Ritter et al., 2012).

2.4. Measurements

Ground-based Swiss measurements for the validation of the model output are available through the National Air Pollution Monitoring Network (NABEL) in hourly resolution for 16 stations (SAEFL, 2003b; EMPA, 2011). Further measurements are available through several cantonal and regional air quality authorities and are provided by the Swiss Society of the responsible for air pollution control (Cercl'Air). All stations are categorized in different settings (e.g. urban, sub-urban, rural and mountainous), whereas mountainous stations (Jungfraujoch, Arosa) are not included in the validation due to their limited ability in representing a 2x2 km^2. Stations directly beside a freeway were reclassified

into their own category. Measured pollutants of interest were NO_2, O_3 and PM_{10}, but not all stations measured all three throughout the two years (e.g. almost no measurements for PM_{10} in 1991). The locations of all the stations used with valid data for at least a coherent month are indicated in Fig. 1.

Meteorological parameters are measured by the Swiss Federal Office of Meteorology and Climatology within their automatic Swiss measurement network (MeteoSwiss, 2004). For 2002, there were 87 valid stations for temperature and 67 for solar radiation. For 1991, there were 68 stations for temperature and 65 stations for solar radiation. The stations are uniformly distributed over Switzerland (see MeteoSwiss (2004)).

2.5. Statistical values

There are a number of statistical parameters that can be used to validate model output. The used parameters for this study were the Pearson correlation coefficient (r), the mean bias (MB), the root mean square error (RMSE) and the mean absolute gross error (MAGE). These parameters are often used and suggested in various scientific literature (Seinfeld and Pandis, 2006; Jacobson, 2005). The formulas below show the equations with OBS being the observations and MOD their corresponding modeled grid points. The number of stations is represented by m and each station has n measurements.

$$MB = \frac{\sum_{i=1}^{n}\sum_{j=1}^{m} MOD_{i,j} - OBS_{i,j}}{nm}$$

$$MAGE = \frac{\sum_{i=1}^{n}\sum_{j=1}^{m} |MOD_{i,j} - OBS_{i,j}|}{nm}$$

$$RMSE = \sqrt{\frac{\sum_{i=1}^{n}\sum_{j=1}^{m}(MOD_{i,j} - OBS_{i,j})^2}{nm}}$$

$$r = \frac{\sum_{i=1}^{n}\sum_{j=1}^{m}(MOD_{i,j} - \overline{MOD}_{i,j})(OBS_{i,j} - \overline{OBS}_{i,j})}{\sqrt{\sum_{i=1}^{n}\sum_{j=1}^{m}(MOD_{i,j} - \overline{MOD}_{i,j})^2}\sqrt{\sum_{i=1}^{n}\sum_{j=1}^{m}(OBS_{i,j} - \overline{OBS}_{i,j})^2}}$$

3. Post-processing

Air quality models are likely to have systematic model errors. On the one hand this is because they rely on emissions estimates along with their uncertainties and on the other hand because of the limitations of the physical and chemical mechanism and their interaction. First results showed an overall systematic bias for O_3 and NO_2 by WRF-Chem. The modeling errors have a seasonal effect and also vary strongly by type of location (rural, sub-urban, urban, freeway) of the measurement station. Recently, Sicardi et al. (2012); Borrego et al. (2011); Djalalova et al. (2010) showed the importance of post-processing of raw model output. Methods such as Bias-correction, Kalman-Filter and Model output statistics (MOS) are already widely used for meteorological forecasting and are more and more applied to air quality forecasting. For this study, we tested different versions of a bias-correction and multi-linear regressions. The predictors for the multi-linear regression were the raw model output, meteorological parameters (temperature, solar radiation, planetary boundary layer height, wind vectors and humidity) and modified sine and cosine functions representing the seasonal variation. These predictors were chosen in prior tests by a forward stepwise selection procedure. Both methods were first applied to all stations combined. This approach managed to improve the seasonal error, but the bias per category of the location was still significant. Best results were achieved with a multi-linear regression for each station, but the derived correlation factors are just applicable to one particular station. Therefore, we calculated a multi-linear regression separately for each category (rural, sub-urban, urban, freeway). Only 50% of the stations were used for the calculation of the correlation factors. These stations were selected randomly if they were not too close to each other. The other half of the stations serve for the validation of the applied post-processing method.

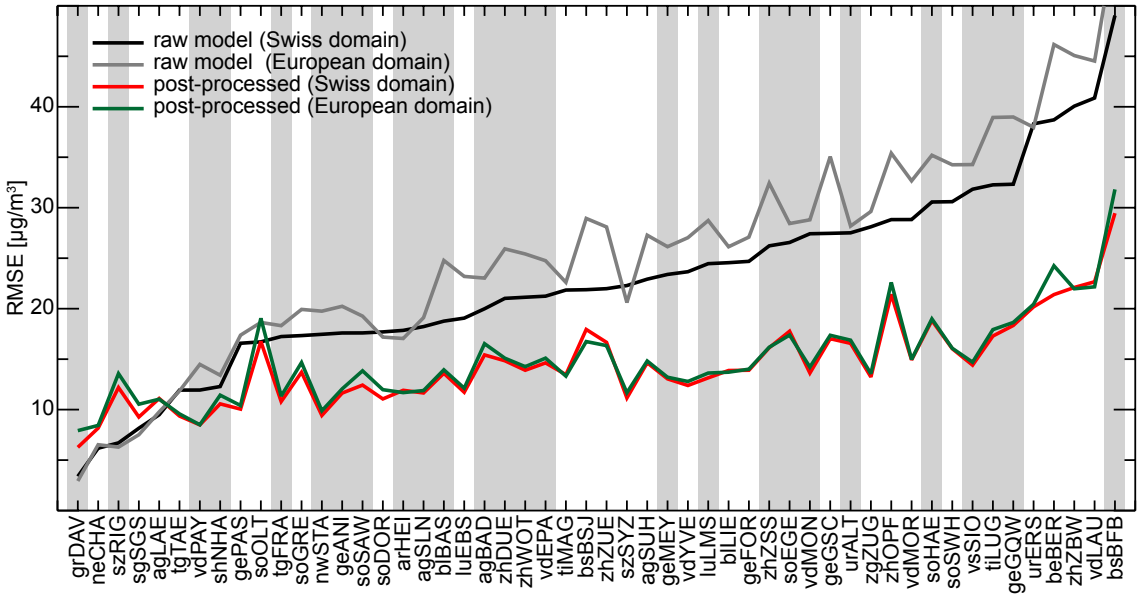

Figure 2: NO_2 root mean square errors for the raw model output and post-processed output (mulit-linear regression) for the Swiss and European domain (year: 2002). Stations without a gray background served for validation.

Fig. 2 shows the RMSE for NO_2 for the year 2002. The stations were sorted according to the RMSE of the raw model output. The station with gray background are the stations which were used to get the correlation factors. For over 90% of the stations, a significant improvement is achieved. The higher the RMSE of the raw model output is, the higher the gain from post-processing. Fig. 2 also reveals that the raw model output of the high resolution domain (Switzerland) performs slightly better than the coarser European domain. If we train the multi-linear regression with raw model output from the coarser European domain and high resolution meteorological data, we achieve almost the same results. This can be useful for operational meteorological forecasts, which can be paired with a coarser chemical transport model.

The absolute differences of NO_2 RMSE values of the raw model output and NO_2 RMSE values of the post-processed data are shown in Fig. 3. The stations are sorted by the absolute differences and the stations with a gray background are the same stations which were used for calculating the correlation factors. The improvement of the validation stations are almost as good as the improvements for the trained stations. No clear pattern can be seen between the stations with a gray background and the stations without. The mean of the absolute differences of the RMSE for the validation station is only increased by 1.02 $\mu g/m^3$ compared to the trained stations. The pattern looks the same for O_3 as well as for PM_{10}.

As the aim is to have the best available post-processed model output for the Swiss domain we decided to take all the stations into account for the correlation factors. The location categories for the whole domain were developed with the official federal land-use categories and other datasets of the Swiss Federal Statistical Office. All results presented below (aside from the meteorological ones) are derived from the post-processed model output with a multi-linear regression per location category trained on all available measurements.

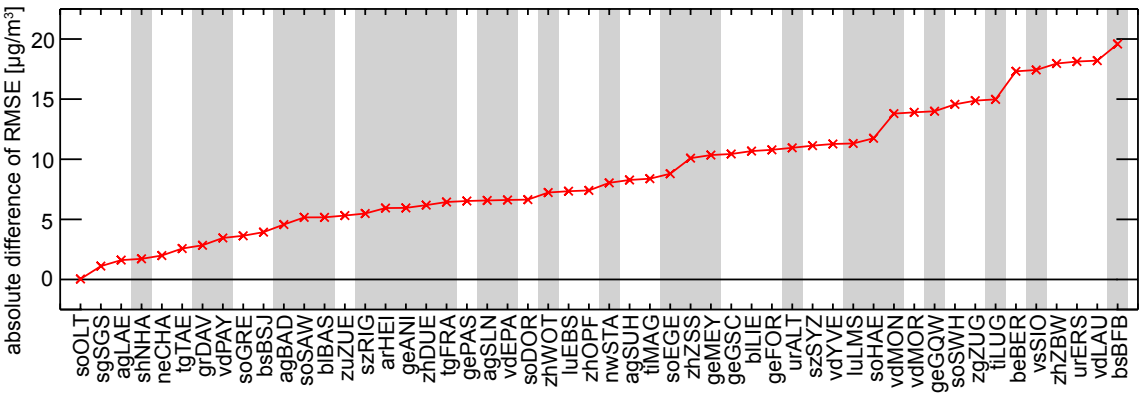

Figure 3: NO_2 absolute difference of root mean square error of raw model output and post-processed output for 2002 (Swiss domain). Stations without a gray background served for validation.

4. Results

4.1. Evaluation of meteorological parameters

We compared the 2 m above ground temperature and the solar radiation model output to measurements operated by the Swiss Federal Office of Meteorology and Climatology. We focused on these two parameters as they are important for most air pollutants. Fig. 4A shows daily averaged values for the mean of all stations and their corresponding modeled grid points for the year 2002. No post-processing was applied to the meteorological parameters, so that these modeled results are from the raw model output. The absolute difference of temperature between the model and the measurements is roughly around 2.5 K in all of 2002. Seasonal trends are well captured and there is good overall agreement. There are only some slightly larger discrepancies for the first half of January and the second half of December. These discrepancies are reflected in all tested air pollutants. The statistical parameters for the temperature can be found in the upper part of Tab. 2. The Pearson correlation coefficients for the years 2002 and 1991 are both 0.92 and the root mean square error is 3.30 for 2002 and 3.51 for 1991, respectively. Müller (2011) found similar results over complex terrain without any post-processing.

Solar radiation is also well correlated between modeled and measured data in Fig. 4B. We have to take into account, that Fig. 4B shows daily mean values and therefore include night hours with little or no solar radiation. Almost all trends are captured by the model. Only some negative peaks of the observations could not be modeled (e.g. mid June) due to non-captured clouds by the model. The absolute difference is lower than 5 W/m^2 in winter and goes up to around 20 W/m^2 in summer. Pearson correlation coefficient of 0.88 and 0.89 for 2002 and 1991 could be achieved (see Tab. 2).

4.2. Evaluation with measurements

4.2.1. Ozone

Daily average values summarized over all stations, their corresponding model grid point, and their absolute difference can be seen in Fig. 5A. The seasonal variation is captured and there is an overall mean absolute difference of around 15 μg/m^3. The biggest differences can be found for the first half of January and the second half of December. These periods are also the ones where the model has the biggest error in predicting temperature. The differences are mainly due to stations classified as urban and freeway. The modeled seasonal trend has smaller peaks than the measured one, which is normal as the model represents an area of 2x2 km^2 and not only a point location. The diurnal cycle has a smaller amplitude for the modeled results. In particular, the daily maximum values are often not modeled correctly, therefore, the model is not able to satisfactorily predict threshold values of air quality standards (e.g. 120 μg/m^3 1-h O_3 mean for Switzerland). The statistical values in Tab. 3 show, that Pearson correlation coefficients for daily maximum values (0.73) are slightly better than daily averaged values (0.67) for 2002, but the RMSE values are

Table 2: Seasonal statistical values of the validation of meteorological parameters (temperature, solar radiation).

Parameter	Year	Season	MB	MAGE	RMSE	r
T 2m	2002		0.13	2.59	3.30	0.92
		DJF	0.80	2.80	3.64	0.81
		MAM	-0.18	2.63	3.30	0.89
		JJA	-0.47	2.43	3.13	0.88
		SON	0.39	2.50	3.13	0.88
	1991		-0.05	2.73	3.51	0.92
		DJF	0.86	3.27	4.25	0.79
		MAM	-0.56	2.56	3.21	0.88
		JJA	-0.81	2.47	3.11	0.90
		SON	0.34	2.63	3.33	0.90
SR down	2002		16.59	55.62	122.93	0.88
		DJF	-2.78	25.75	58.13	0.86
		MAM	23.56	68.63	141.03	0.87
		JJA	42.30	83.40	166.73	0.87
		SON	2.69	43.94	95.17	0.85
	1991		18.09	52.85	116.35	0.89
		DJF	0.57	24.56	56.32	0.90
		MAM	25.78	69.50	138.53	0.88
		JJA	41.77	76.83	155.42	0.89
		SON	3.89	39.99	87.30	0.88

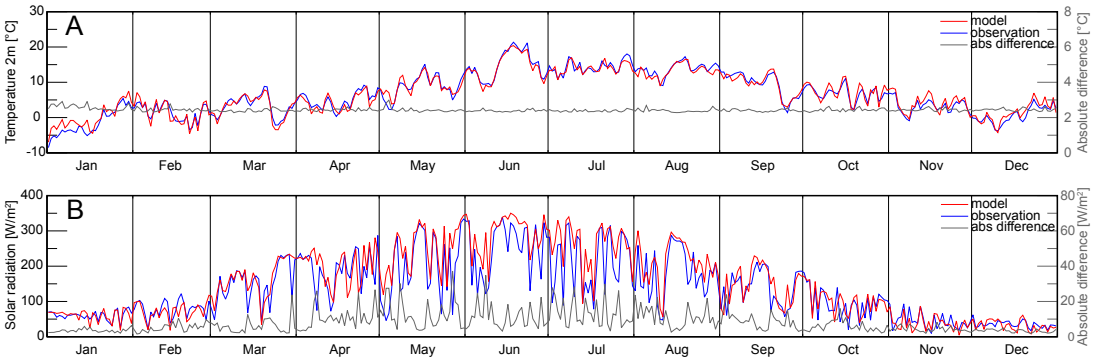

Figure 4: Daily averaged values for the mean of all stations, their corresponding grid points and the absolute difference of them for temperature and solar radiation (year: 2002).

also increased. Tab. 3 also shows hourly averaged values performing slightly worse (0.53). Baldasano et al. (2011); Schürmann et al. (2009) presented similar correlations for high resolution domains of Spain and Southern Italy, respectively. If we look at yearly statistics (see Tab. 4) the Pearson correlation coefficient is lower with 0.53 and 0.68 for 2002 and 1991, but RMSE and MAGE values are very low.

Fig. 6A and Fig. 7A show the annual average concentration and the difference of the measurements with the corresponding grid point for the year 2002. We see low ozone levels over cities and high ozone levels over the mountains. The former phenomenon is due to high nitrogen oxides concentration in cities. The latter phenomenon comes from a dependence of O_3 on altitude. If we look at Fig. 7A, we see that there are only two border region that can be distinguished. The first is the region around Geneva which seems to overestimate modeled values and the second

is the region of Basel which underestimates modeled values. The two highest negative differences are from stations with an altitude higher than 1000 m. The two most positive differences are from stations with high traffic volume, but not classified as freeway stations. These stations have more nitrogen oxides due to traffic and therefore less ozone than predicted.

Table 3: Statistical values of the validation of O_3, NO_2 and PM_{10} for hourly averaged, daily averaged and daily maximum values.

Pollutant	Year	category	MB	MAGE	RMSE	r
O_3	2002	hourly	4.76	25.02	30.63	0.53
		daily	4.71	18.85	21.91	0.67
		daily max	1.90	21.29	27.25	0.73
	1991	hourly	4.43	25.52	32.44	0.65
		daily	4.35	18.67	23.10	0.75
		daily max	0.72	27.93	36.85	0.74
NO_2	2002	hourly	-2.96	13.72	18.02	0.38
		daily	-2.93	10.11	13.04	0.54
		daily max	-6.04	16.45	20.86	0.51
	1991	hourly	-4.69	18.79	25.39	0.50
		daily	-4.70	14.03	19.03	0.65
		daily max	-11.67	23.18	31.52	0.61
PM_{10}	2002	hourly	-0.78	13.98	19.92	0.12
		daily	-0.83	10.34	14.68	0.22
		daily max	5.11	20.15	30.25	0.13
	1991	hourly	-0.95	17.25	23.61	0.32
		daily	-0.98	15.34	21.44	0.44
		daily max	14.65	23.72	27.71	0.39

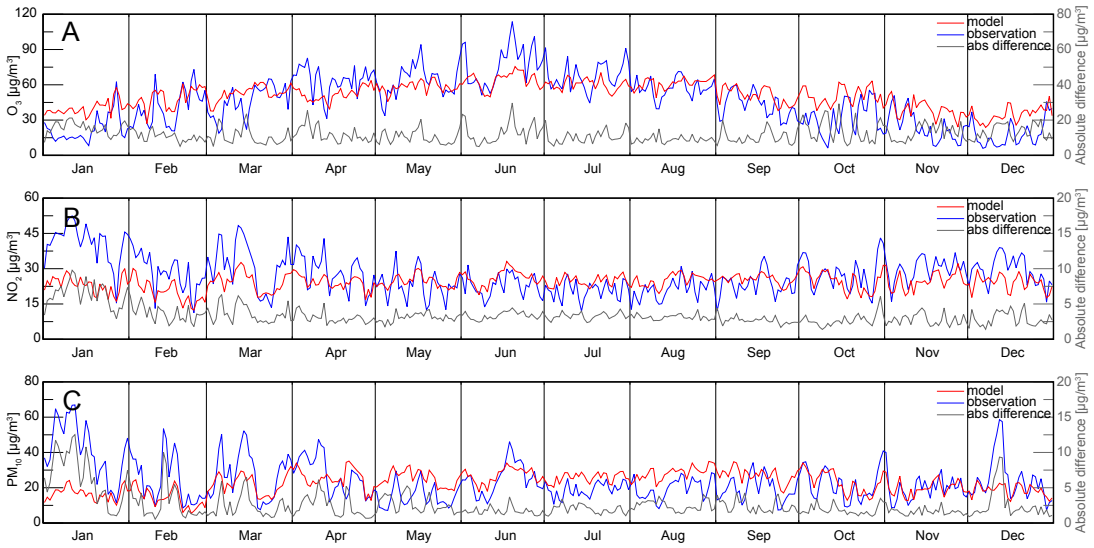

Figure 5: Daily averaged values for the mean of all stations, their corresponding grid points and the absolute difference of them for O_3, NO_2 and PM_{10} (year: 2002).

4.2.2. Nitrogen dioxide

Looking at the daily average curve for the mean of the observations (Fig. 5B), modeled NO_2 values are in good agreement with measurements. The absolute difference generally varies between 2-5 $\mu g/m^3$ except for the first two months where we have a difference of up to 10 $\mu g/m^3$. Note that the same effect could be seen for ozone and temperature. These discrepancies exist for all categories but are most significant for freeway locations. Overall, the corresponding grid points for freeway stations predict very low NO_2 concentrations. For rural stations the model predicted about 2 $\mu g/m^3$ too much for summer months. Aside from winter months where the model data are too low (see Fig. 5B, diurnal cycles are well captured. Minimum and especially maximum peaks are not well represented by the model. Tab. 3 confirms this fact with Pearson correlation coefficient that are lower for daily maximum values than for daily averaged values. If we summarize the data to yearly values, we reach Pearson correlation coefficients of 0.72 and 0.80 for 2002 and 1991 respectively (see Tab. 4).

A look at the yearly averaged map for 2002 in Fig. 6B reveals almost the opposite pattern as for O_3. Cities and their surroundings have very high NO_2 concentrations and, the higher the altitude, the lower the nitrogen oxide emissions, hence NO_2 concentrations are lower. Some major freeways in Switzerland can be identified by increased NO_2 values. For most of the measurement stations the model calculated very low concentrations (see Fig. 7B). Stations in steep valleys and urban canyons have the biggest differences. All stations with high negative differences are near roads with high traffic density. It seems difficult to represent stations in steep valleys and urban canyons with a horizontal resolution of 2x2 km^2. NO_x emissions are probably underestimated for these specific locations as all sub-grid emissions are just summarized into the same grid cell.

4.2.3. Particulate matter

We concentrated the validation of particulate matter on PM_{10} in 2002 due to the limited number of $PM_{2.5}$ measurements in 2002 and 1991, and the smaller number of PM_{10} measurements in 1991. Almost no seasonal (see Fig. 5) or diurnal cycle can be observed. However, the observations show high random daily peak values of over 60 $\mu g/m^3$. These high peaks are not well captured by the model and are mainly due to rural and freeway stations (which are mostly in rural environments). One of these high peaks occurred in mid December. Aside from the special situation in January, this is one of the only situations where an absolute difference of more than 7.5 $\mu g/m^3$ was observed. In general, the diurnal cycle underestimates PM_{10} concentrations in winter and overestimates them in summer months. We noticed low Pearson correlation coefficient for hourly averaged, daily averaged and daily maximum comparisons (see Tab. 3). Nevertheless, summarized over the entire year, we achieved a reasonable Pearson correlation coefficient of 0.71 for 2002 (see Tab. 4).

Fig. 6C shows an altitude dependence of PM_{10} with lower concentrations in higher regions. Cities and their surroundings have higher concentrations. We observe average values in rural regions which are due to a variety of emission sources including natural, non-anthropogenic ones. Bigger valleys have relatively higher concentrations just like big lakes (e.g. Lake Geneva and Lake Constance). The most interesting part is the region around canton Ticino and the northern part of Italy. Although this region has no extremely high emissions compared with the northern part of the Alps, the model predicts very high annual concentrations of up to 30 $\mu g/m^3$. However, a quick look at the coarser European domain clearly indicates that the whole Po Valley has increased values due to heavy industrial cities like Milan and Turin. The concentrations around Lugano are thus due to advection from the coarser European domain (Southern border). Grell et al. (2000) showed a similar advection scheme for this region. Even though the modeled values for Ticino were so high, the differences between the model and the measurements are still negative. The two biggest positive differences are on the Rigi (over 1000 m altitude) and in the city of Olten. Most negative differences, aside from those in Ticino, are at freeway locations or places with very high traffic density. The biggest difference of around -12 $\mu g/m^3$ is in a steep street canyon in the capital of Switzerland (Bern).

4.3. Comparison with PolluMap

We also compared the model output to an already existing dispersion modeling system (PolluMap version 2.0). PolluMap is an empirical model which uses transfer functions to represent the impact of emission sources on neighboring areas. Calculations were done for yearly averaged PM_{10} and $PM_{2.5}$ as well as NO_x with a conversion function to NO_2 for the years 1990 and 2000 (SAEFL, 2003a, 2004b; Liu et al., 2007). PolluMap was run with the same emissions as those used in this study for PM_{10} and NO_2 at a horizontal resolution of 200 m. In addition to these transfer

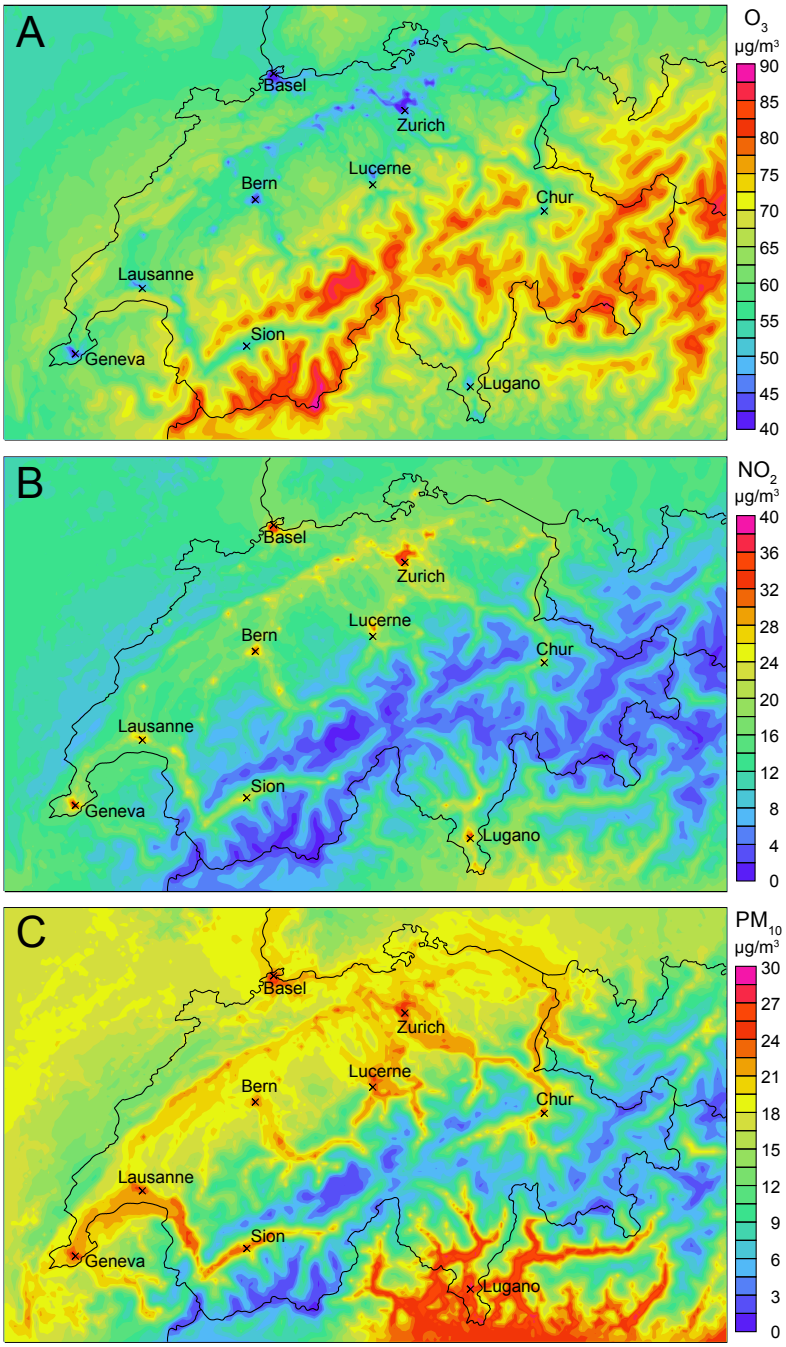

Figure 6: Annual mean concentrations for O_3, NO_2 and PM_{10} for the year 2002.

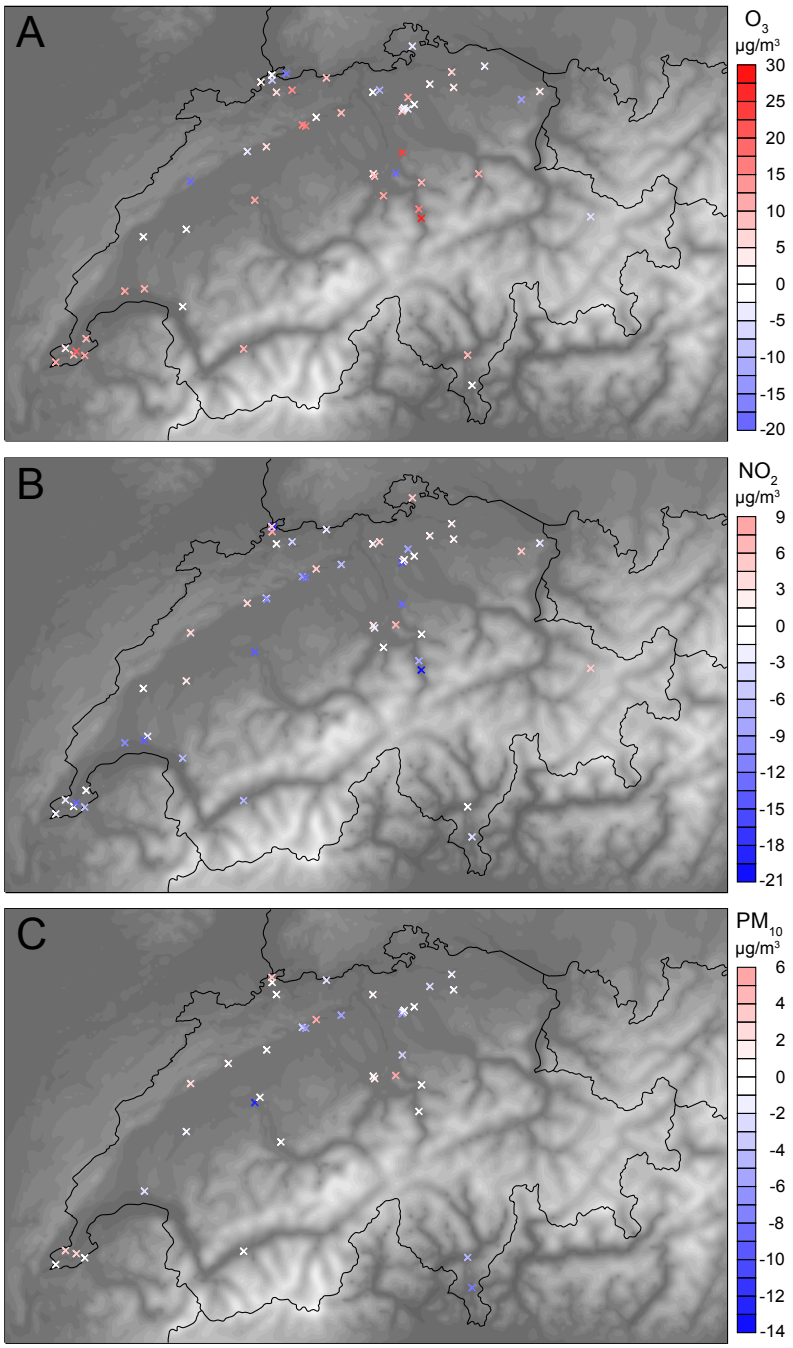

Figure 7: Mean of hourly differences between model output and observations for O_3, NO_2 and PM_{10} (year: 2002).

functions, the model adds a rural-urban transfer functions and spatial background concentrations which were trained by measurements. It is important to keep in mind that we are comparing completely different modeling approaches as well as different years. We compare only yearly averaged model output data (WRF-Chem and PolluMap) to measurements as only these datasets were available from PolluMap. Fig. 8 shows a scatterplot of modeled vs. measured data for NO_2 and PM_{10} for the year 2002 and 2000, respectively. The corresponding statistical values are listed in Tab. 4. Both models have roughly the same statistical values for PM_{10}. It seems that PolluMap performs slightly better for NO_2. A possible explanation could be the better correlation of the PolluMap output with the original emissions (r=0.52), compared to the WRF-Chem output (r=0.45). Nevertheless, WRF-Chem is producing three dimensional data for a variety of pollutants, whereas PolluMap is restricted to ground-based results and just particulate matter and nitrogen oxides. Furthermore WRF-Chem is capable of producing valid daily output as Tab. 3 shows.

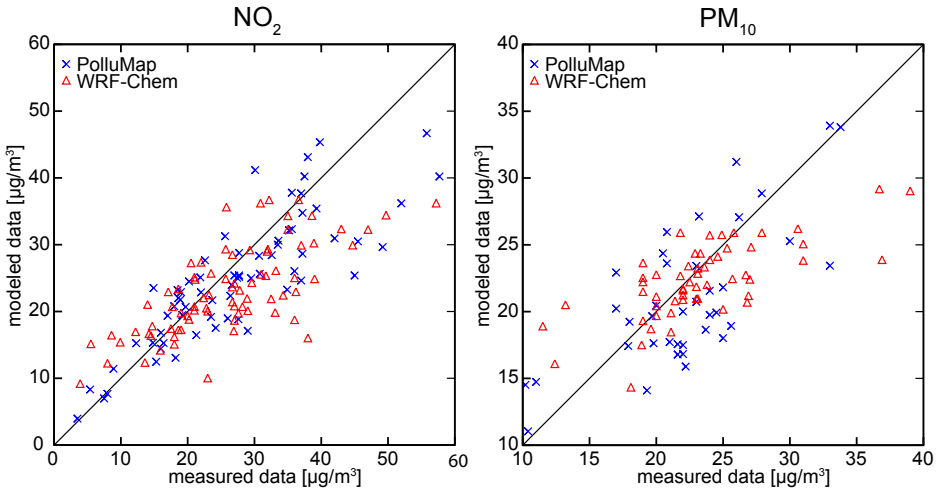

Figure 8: Scatterplot of modeled and measured data for NO_2 and PM_{10} for the year 2002 (WRF-Chem) and 2000 (PolluMap).

Table 4: Yearly statistical values of the validation of O_3, NO_2 and PM_{10} for WRF-Chem and PolluMap.

Pollutant	Model	Year	MB	MAGE	RMSE	r
O_3	WRF-Chem	2002	5.32	10.90	13.45	0.53
		1991	6.25	10.95	13.38	0.68
	PolluMap	2000	-	-	-	-
		1990	-	-	-	-
NO_2	WRF-Chem	2002	-3.00	5.92	7.70	0.72
		1991	-5.01	8.00	10.38	0.80
	PolluMap	2000	-2.54	5.15	6.95	0.83
		1990	-6.02	7.36	10.06	0.86
PM_{10}	WRF-Chem	2002	-1.00	2.74	3.88	0.71
		1991	-	-	-	-
	PolluMap	2000	-1.12	3.46	4.11	0.72
		1990	-	-	-	-

5. Discussions and Conclusions

This paper presented an evaluation of the online meteorology-chemistry model WRF-Chem over complex terrain (e.g. Switzerland) for two full years. We focused the results on the temporal and spatial distribution of pollutants compared to measurements and an already existing empirical dispersion model. The raw model output has been post-processed with a multi-linear regression for different categories (rural, sub-urban, urban, freeway). For over 90% of all corresponding grid points of the measurements, the RMSE could be reduced, even though only half of all stations served for the calculation of the regression factors. We noticed that the raw model output of the high resolution Swiss domain preformed slightly better than the raw model output of the coarser European domain. However, if we post-process with air pollutants of the coarser domain and high resolution meteorological data, we achieve almost the same result as if we train the routine with only high resolution data. As the computational effort between a run with meteorology and chemistry together versus only a meteorological run is about a factor of 10, we conclude that the best approach with respect to computational time would be a coarser domain with the calculation of the pollutants and a nested domain with just meteorological forecasting. This approach could be used to establish an air pollution forecast system.

The model performed well for meteorological parameters as the temperature had only a mean bias of 0.13 and -0.05 for 2002 and 1991 with Pearson correlation coefficients of 0.92. The results of the solar radiation had r-values of 0.88 and 0.89 for 2002 and 1991 respectively. Only the month of January had a situation that was not captured properly by the model, this same effect was also seen for temperature and air pollutants. Correlation factors for the multi-linear regression showed that temperature had significant influence on air pollutants (especially for ozone).

Daily averaged values of the mean of all stations and their corresponding grid points for 2002 showed that seasonal variations are represented correctly for O_3, NO_2 and PM_{10}. However, peak values could not be reproduced by the model but, as the model only has grid cells of 2x2 km^2, the results seem acceptable. Clearly this kind of model cannot be used for regulatory purposes with regard to air quality standards as set by the European commission and the Swiss government. Special post-processing routines for maximum values would be necessary to replicate these standards. For O_3, the best Pearson correlation coefficient was achieved with daily mean values; whereas, annual statistics lead to the best results for NO_2 and PM_{10}. Spatial annual patterns look reasonable for all three pollutants. Ozone and NO_2 seemed to have opposite patterns but this seems reasonable as nitrogen oxide and nitrogen dioxide are precursors for the ozone formation. The PM_{10} distribution shows the advection of particulate matter from the Po valley to southern parts of Switzerland (e.g. Ticino). Regarding the spatial error distribution, we see almost no clear pattern except for border regions like areas around Geneva and Basel. Overall, most of the stations have only small differences; however, some bigger differences can be explained with the special characteristics of the location of the station (e.g. on mountains, in urban street canyons or beside heavy traffic roads). The model was particularly good for rural areas but could also reproduce background levels in urban areas.

The comparison with PolluMap shows that WRF-Chem can produce results that are as good as a specially dedicated dispersion model. In addition, WRF-Chem produces daily mean values that are satisfactory (especially for O_3) in contrast to only yearly averages from PolluMap. Furthermore, WRF-Chem calculates a three-dimensional grid for a variety of air pollutants and meteorological parameters. Disadvantages of the physical-chemical modeling approach are requirements of extensive computing resources on the hardware-side and the high level of system skills on the software-side. On the other hand, new modules such as a secondary organic aerosol (SOA) module could easily be implemented into an already existing modeling system. Next steps could be to look at other health relevant pollutants (e.g ultrafine particles, sulfur dioxide) and to couple the model results with land-use regression models for some limited highly populated areas to achieve an even higher resolution. WRF-Chem and similar models can provide highly time resolved output over large areas for many pollutants. Such data can richly add to air pollution health research.

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3.3. Spatio-temporal differences and diurnal cycles

This subsection is a further analysis of the Swiss domain results. It shows modeled pollutants for 2010, their spatio-temporal difference between 2002 and diurnal cycles. The pollutants analyzed are O_3, NO_2, NO, PM_{10} and $PM_{2.5}$. Spatio-temporal differences between the years 1991 and 2002 or 1991 and 2010 are not shown because the spatial pattern of the years 1991 and 2002 are pretty much the same as they have the same emission basis. However, 2010 has a completely new emission basis including point sources with very high, local emissions. It should also be remembered that meteorological IC/BC's are not the same for these two modeled years. Reanalysis II data are used for 2002 and GFS data for 2010, respectively. Nevertheless, these meteorological IC/BC's are only used for the European domain as the Swiss domain is directly nested into the European one. Maps of yearly averages for O_3, NO_2 and PM_{10} for the year 2002 are already shown in Fig. A2-6.

3.3.1. Ozone - O_3

Post-processed O_3 Ozone concentrations are strongly dependent on temperature and its precursors (e.g. NO_2). This fact is already shown in Chap. 3.2 as temperature had very high correlation factors for the multi-linear regression and opposite spatial pattern to NO_2. Therefore, ozone concentrations in regions with very high NO_x emissions (e.g. cities) have low values and alpine regions with low concentrations and low temperature have relatively high O_3 concentrations. Fig. 16A shows the annual mean for the year 2010. The biggest cities (Zurich, Geneva, Basel) have concentrations around 40-50 $\mu g/m^3$ whereas mountainous areas have concentrations up to 85 $\mu g/m^3$. Over the lakes, higher values can be seen as over their direct surroundings. This comes from the influence of temperature through multi-linear regression. Fig. 17A shows the yearly averages of temperature 2 m above ground. In general, the lakes in 2010 have the highest temperature. Water acts as a huge heat storage reservoir and emits the heat from daytime during the colder hours.

If yearly averages between 2010 and 2002 are compared (see Fig. 16B) up to 6 $\mu g/m^3$ can be seen over lakes. A look at Fig. 17B shows almost no differences in temperature between these two years over the lakes. On the other hand it can be seen that the year 2002 was about 2 °C warmer than 2010. The temperature difference between the lakes and their surroundings is consequently bigger in 2010 than it was in 2002. This leads to the higher ozone concentrations in 2010 over the lakes compared to 2002. Most of the alpine regions have a small decrease of O_3 whereas the Swiss Plateau and the Ticino have slightly increased O_3 values. Overall, the changes are rather small over land. There are some negative differences of up to 6 $\mu g/m^3$ (e.g. north-west of Zurich and north-west of Bern). They are due to local NO_x point sources. As they were not implemented in the emissions of 2002, they appear as differences between 2010 and 2002.

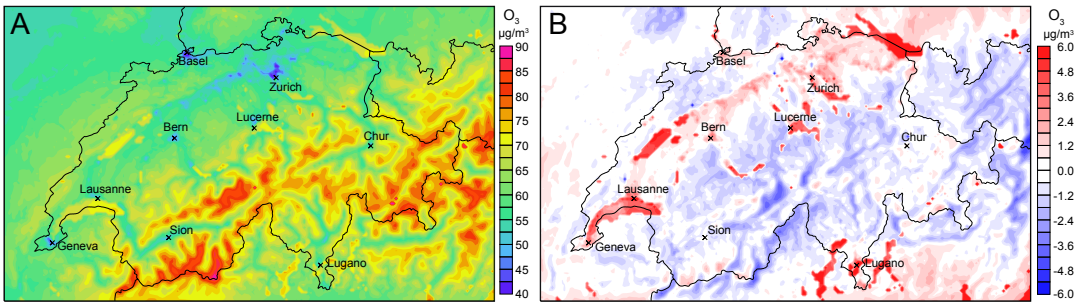

Figure 16: Yearly averages of ozone concentrations for the year 2010 (A) and its spatio-temporal difference to the year 2002 (B). The concentrations have been post-processed by a multi-linear regression algorithm.

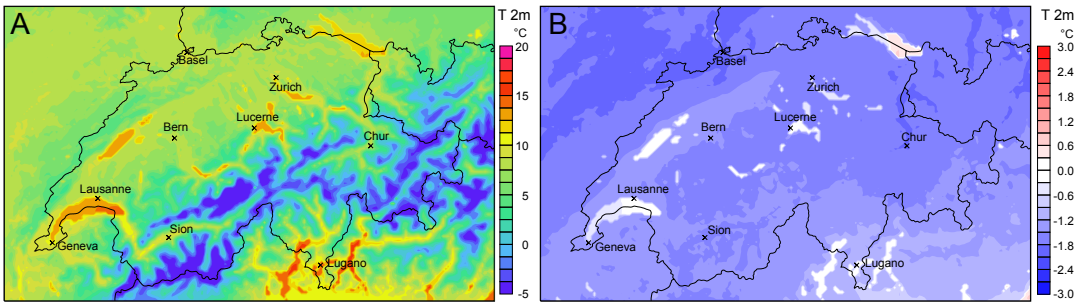

Figure 17: Yearly averages of temperature (2 m above ground) for the year 2010 (A) and its spatio-temporal difference to the year 2002 (B).

Raw model output O_3 Not all pollutants have sufficient measurements for the applied post-processing method. Therefore, it was tested to see if the bias of the raw model output is approximately the same for both years so that the spatio-temporal differences of such pollutants would be the same for post-processed as raw model results. Fig. 18 shows the yearly averages of O_3 concentrations and the differences to 2002 for the raw model output. Compared to Fig. 16A there is much less spatial details. However, very low concentrations can still be seen at local NO_x point sources and high concentrations in mountainous regions. Overall, the raw model output predicts concentrations in alpine areas that are too high. If one looks at the differences to the year 2002 for the raw model output, the ozone concentrations decreased over these eight years for almost the whole of Switzerland. Some patterns of the post-processed results can still be observed. For example the region south-east of the Ticino has still slightly increased values and the Swiss Plateau has smaller differences than other regions. However, a lot of spatial information is added by the multi-linear regression. Unfortunately, spatio-temporal biases are different for these two years. Changes in the main pattern can also be seen in the raw model output but detailed maps and differences can only be achieved for O_3 by applying the post-processing routines.

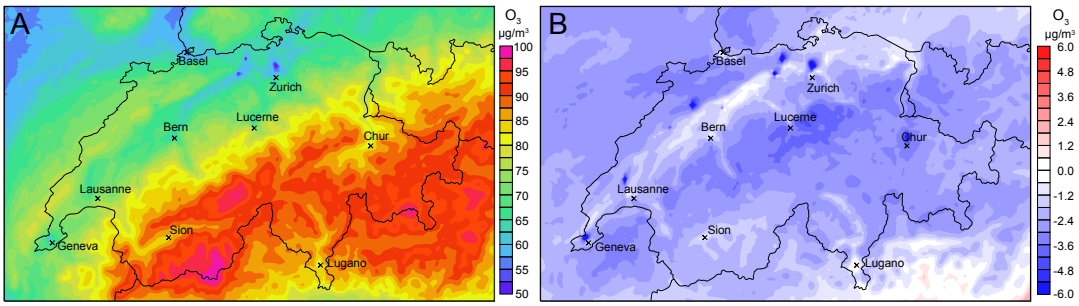

Figure 18: Yearly averages of ozone concentrations for the year 2010 (A) and its spatio-temporal difference to the year 2002 (B). The concentrations originate from the raw model output without any post-processing.

Diurnal cycle O_3 A look at the diurnal patter reveals no big differences between the two years (see Fig. 19). Overall, the dashed lines of 2002 have slightly lower concentrations than the ones for 2010. It must be remembered that only the corresponding grid points of the observations are summarized into this figure. Most of these stations are in the Swiss Plateau where Fig. 16B shows an increase of O_3 from 2002 to 2010. The model has a smaller amplitude and predicts the concentration peak of the afternoon to early. The negative peak in the morning hours around 7 o'clock is not captured by the model. Winter months (December, January, February) have overall a smaller amplitude and lower values. On the other hand summer months (June, July, August) have the biggest amplitudes for the measurements and the model. Spring months (March, April, May) and fall months (September, October, November) are between the patterns of summer and winter months.

3.3.2. Nitrogen oxide - NO_2 & NO

Post-processed NO_2 Nitrogen dioxide often has opposite spatial patterns than O_3. Anthropogenic emissions are mainly due to road traffic and point sources. Fig. 20A shows the yearly mean of 2010. The mountainous areas have very low concentrations below 10 $\mu g/m^3$. This is due to the lack of primary emissions. On the other hand relatively high mean values can be seen in the biggest cities (Zurich, Geneva and Basel), around some industry point sources and following freeways and major roads. As the freeways and the major roads are often in valleys (e.g. Rhône valley) the NO_2 concentrations are spread into the whole valley. Overall, the region around Ticino has slightly increased values. The influence of temperature on NO_2 is not as strong as for O_3, otherwise the post-processing routines would have made the Swiss lakes in Fig. 20A visible.

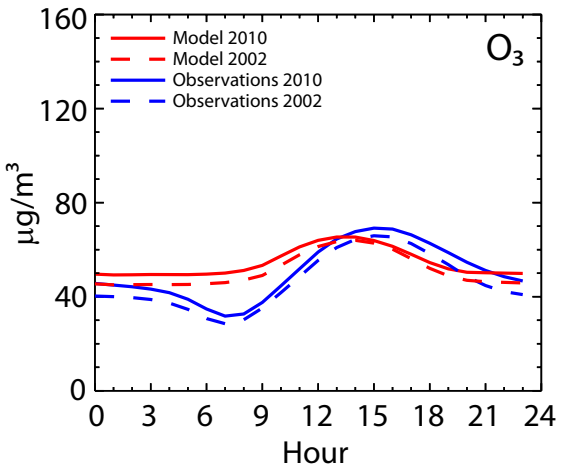

Figure 19: Averaged diurnal cycles for O_3 for 2010 and 2002. The mean of all available stations and their corresponding model grid points is shown.

Even though nitrogen oxide emissions decreased more than 20% during the last decade, Fig. 20B shows an overall increase of NO_2 concentrations about 3 $\mu g/m^3$. Especially alpine regions have increased values, whereas some part of northern Italy have a small decrease. Big valleys and the Swiss Plateau have no or only a small increase of NO_2 concentrations. As point sources were absent for modeling the year 2002, small regions with high differences can be seen. The concentrations are dispersed to some degree from the point sources.

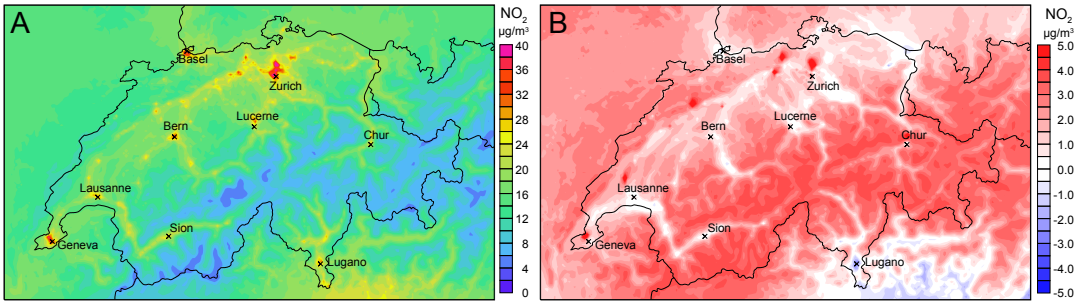

Figure 20: Yearly averages of NO_2 concentrations for the year 2010 (A) and its spatio-temporal difference to the year 2002 (B). The concentrations have been post-processed by a multi-linear regression algorithm.

Raw model output NO_2 As for ozone, the raw model NO_2 concentrations have less spatial details than the ones that were post-processed. The annual mean and the difference to 2002 can be seen in Fig. 21. Figure A has another scale than Fig. 20A. The model shows almost no NO_2 concentrations over the Alps and only few (around 10 $\mu g/m^3$) in the Swiss Plateau and around 20 $\mu g/m^3$ in the bigger region of Zurich and at the point sources. The model has a big bias and models only half of the actual concentrations. Urban environments are especially poorly represented. This shows how necessary post-processing routines are. However, if the differences are kept in mind, the raw model output actually performs more reasonable than the post-processed data. Measurements from the Federal Office for the Environment (FOEN) show almost no differences for alpine foothills, a slight decrease for rural stations and a decrease for urban areas (see Fig. 29). Fig. 21B shows exactly these criteria. Increased values only exist around the industrial point sources which is reasonable as point sources were not implemented for the 2002 calculation. Nevertheless, the spatial pattern between Fig. 21B and Fig. 20B are almost the same, but with different ranges.

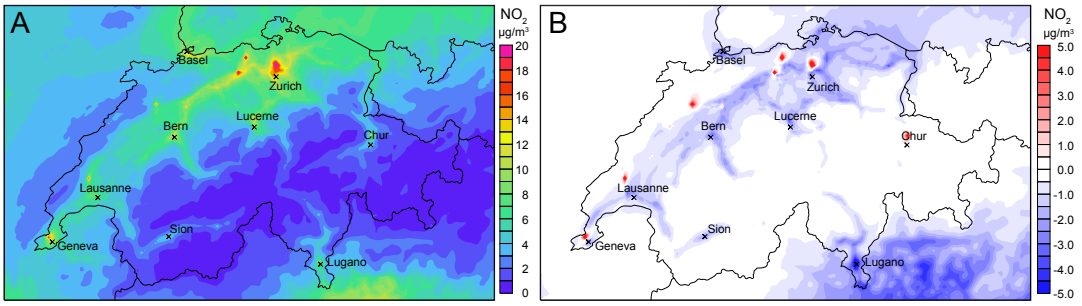

Figure 21: Yearly averages of NO_2 concentrations for the year 2010 (A) and its spatio-temporal difference to the year 2002 (B). The concentrations originate from the raw model output without any post-processing.

Diurnal cycle NO_2 Looking at the diurnal cycles of NO_2, no differences between the two years can be seen (see Fig. 22). As for O_3, most of the stations are situated in the Swiss Plateau were Fig. 20A shows almost no differences. Modeled values are overall still too small. The two positive peaks (8 am and 8 pm) are noticeable in both diurnal cycles, but the amplitude is much smaller for modeled results. The negative peak of the mean of all observations around 4 am is not represented by the model.

Post-processed NO Nitric oxide is a radical and therefore chemically reactive. When exposed to oxygen it is converted to NO_2 (see Tab. 3). Therefore, spatial pattern of NO and NO_2 are directly linked to each other. Anthropogenic emissions are summarized as NO_x and afterwards split up to NO and NO_2. Fig. 23A shows the yearly mean of NO for the year 2010. The spatial patterns are almost exactly the same as in Fig. 20A. However,

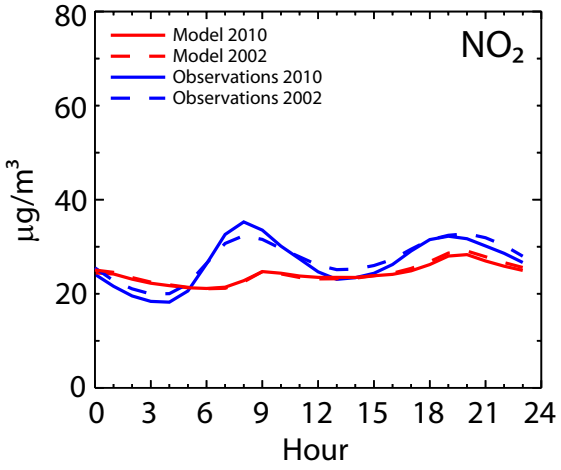

Figure 22: Averaged diurnal cycles for NO_2 for 2010 and 2002. The mean of all available stations and their corresponding model grid points is shown.

overall the range of the concentrations for NO are cut in half. As NO is highly reactive and can not be as far advected as NO_2, industrial point sources and emissions from roads are not spread so far as for NO_2. The raw model output has the same pattern as raw model NO_2. NO concentrations of the raw model output are extremely under-predicted. Overall, predicted NO concentrations are not reasonable without any post-processing.

Differences of NO concentrations between the modeled years 2010 and 2002 are shown in Fig. 23B. Concentrations in valleys have decreased during these years and concentrations on mountains and plains have slightly increased. In contrast to Fig. 20B cities have a relatively high decrease during these years. As the spatial pattern looks almost the same for the NO_2 and NO concentrations for the year 2010, but not the difference maps, the maps for 2002 have to be noncoherent. In fact, the spatial patterns for 2002 for these two pollutants have differences. NO concentrations are relatively higher on freeways. As said before, this is because of the high reactivity of the radical and therefore the decreased capacity to be advected. Anthropogenic NO emissions from roads in 2010 are only 70% of the total emissions from roads for the year 2002. Therefore, the differences are not clearly distinguishable in the spatial pattern of 2010.

Diurnal cycle NO Diurnal cycles for the radical NO shown in Fig. 24 have bigger amplitudes than the one for NO_2. Measurements still have two peaks around 8 am and 8 pm. However, the model is not capable of reproducing these peaks significantly. Overall, the year 2002 has higher values for the observations as well as for the corresponding grid points.

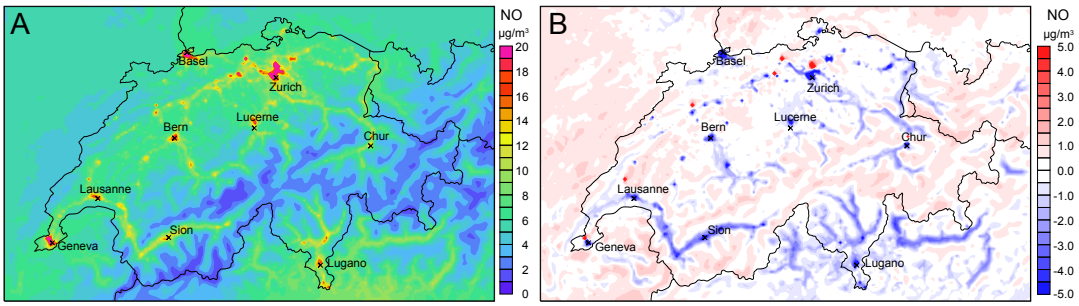

Figure 23: Yearly averages of NO concentrations for the year 2010 (A) and its spatio-temporal difference to the year 2002 (B). The concentrations have been post-processed by a multi-linear regression algorithm.

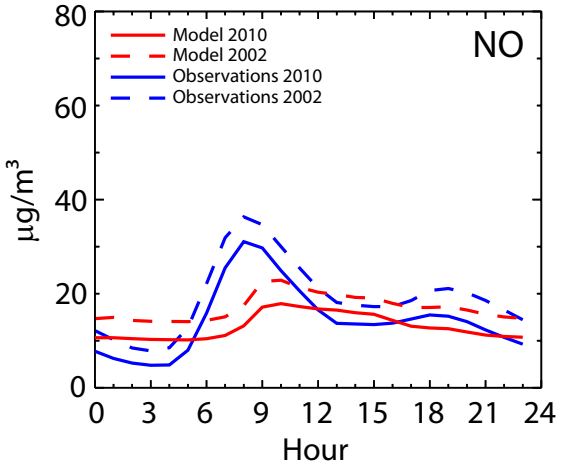

Figure 24: Averaged diurnal cycles for NO for 2010 and 2002. The mean of all available stations and their corresponding model grid points is shown.

3.3.3. Particulate matter - PM_{10} & $PM_{2.5}$

Post-processed PM_{10} Particulate matter is far more complex like O_3, NO_2 or NO. There are many different primary and secondary sources and it can be advected over large distances. However, Fig. 25A shows a clear and distinct pattern. There are high values in the valleys of the canton Ticino and northern Italy, the Swiss Plateau and bigger alpine valleys. Cities also have higher PM_{10} concentrations than their rural surroundings. As mentioned in Chap. 3.2, the highest concentrations in the southern Swiss part are due to advection from high industrial areas in northern Italy.

The differences to the year 2002 reveal an increase over mountainous areas and a decrease in the Swiss Plateau, in Ticino and in big cities (see Fig. 25). The Jura hills have a slight increase of PM_{10} concentrations. There are no significant PM_{10} point sources in Switzerland. Therefore, no differences of local point sources as for NO_2 and NO between exists the years 2002 and 2010.

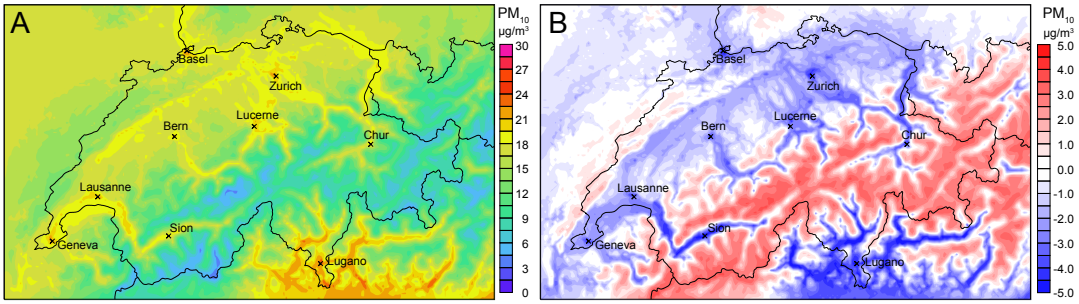

Figure 25: Yearly averages of PM_{10} concentrations for the year 2010 (A) and its spatio-temporal difference to the year 2002 (B). The concentrations have been post-processed by a multi-linear regression algorithm.

Raw model output PM_{10} The influence of post-processing routines on the PM_{10} concentrations is minor compared to the one for the other pollutants. The model performs slightly better after the multi-linear regression, but the raw model output was already in an acceptable range. Therefore no major differences can be seen between Fig. 25A (range 0-30 $\mu g/m^3$) and Fig. 26A (range 0-20 $\mu g/m^3$). However, the post-processed data has more spatial informations due to the separation between different categories (urban, sub-urban, rural, freeway) for the multi-linear regression.

The maps of the differences between the years 2002 and 2010 for PM_{10} differ one from each other. The main spatial pattern remains the same. Alpine regions have no changes in the concentrations (see Fig. 26B). On the other hand, parts of northern Italy and the Swiss Plateau have a decrease around 5 $\mu g/m^3$. Compared to Fig. 30 the differences of the raw model output data matches better the mean of the grouped measurements shown by FOEN (see Fig. 30).

Diurnal cycle PM_{10} There are almost no diurnal cycles for PM_{10} (see Fig. 27. The amplitude for both, observations and corresponding grid points, is very small. Modeled values are in the exactly same range as measurements. In 2002, the values were slightly higher for the model and the observations, which can also be seen in Fig. 26B.

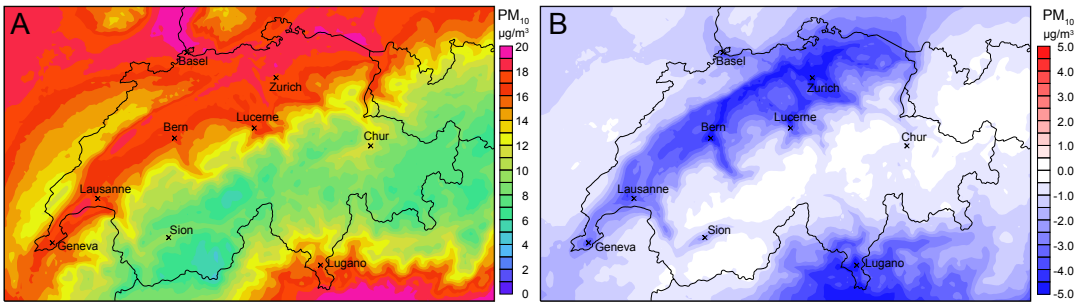

Figure 26: Yearly averages of PM_{10} concentrations for the year 2010 (A) and its spatio-temporal difference to the year 2002 (B). The concentrations originate from the raw model output without any post-processing.

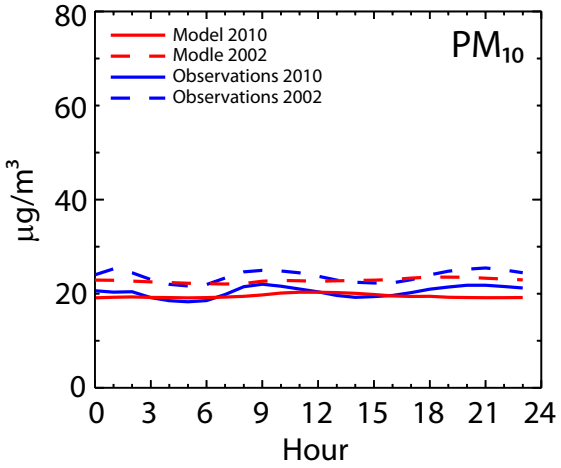

Figure 27: Averaged diurnal cycles for PM_{10} for 2010 and 2002. The mean of all available stations and their corresponding model grid points is shown.

Raw model output $PM_{2.5}$ There were not enough $PM_{2.5}$ measurements to apply a multi-linear regressions as for all other mentioned pollutants. However, the yearly mean of 2010 and the difference to 2002 of the raw model output can be seen in Fig. 28. The spatial patterns are comparable to the same figures for PM_{10} (Fig. 25). $PM_{2.5}$ concentrations are lower than the ones for PM_{10} which is evident, as PM_{10} includes all particle matter smaller than 10 μm (also $PM_{2.5}$).

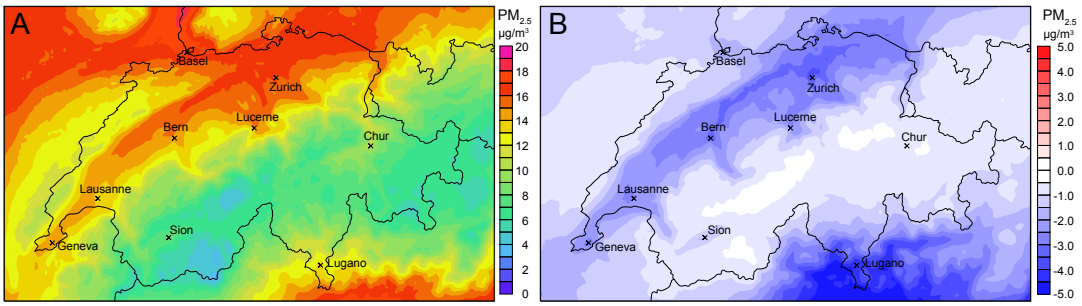

Figure 28: Yearly averages of $PM_{2.5}$ concentrations for the year 2010 (A) and its spatio-temporal difference to the year 2002 (B). The concentrations originate from the raw model output without any post-processing.

3.3.4. Discussions of the spatio-temporal differences and diurnal cycles

Every model is likely to have systematic model errors and WRF-Chem is no exception. The chosen approach manages to eliminate these biases by a mutli-linear regression per category (urban, sub-urban, rural and freeway). The reference values of the post-processing are ground-based point measurements. It is impossible for the available measurement stations to represent the total of an aforementioned category. Looking at one specific year, the post-processing routine improves the modeling result significantly. This is the case for NO, NO_2 and O_3. The improvements for PM_{10} were only small. If post-processed yearly means are compared, another bias is introduced to the difference. If the differences of NO_2 (Fig. 20B and Fig. 21B) and PM_{10} (Fig. 25B and Fig. 26B) are compared to yearly mean of measurements (Fig. 29 and Fig. 30) it can be seen that the raw model output captures the changes over the years better than the post-processed results. This is also applicable for other pollutants. Overall, this means that if the main focus is on air pollution exposure for a specific year or moment, post-processing routines are essential. However, if the main focus is on the changes of pollutants over the years, the raw model output delivers better results.

All diurnal cycles show smaller amplitudes for the mean of the corresponding grid points of WRF-Chem than the mean of the measurements. The more reactive a pollutant is (e.g. NO), the more the amplitudes differ for the model and the measurements. Positive peaks are mainly captured by the model. However, peaks are shifted up to one hour later or earlier. The differences of the diurnal cycles between the years 2010 and 2002 are minor. Both, the model and the measurements show slightly higher values for O_3 and slightly lower values for NO and PM_{10} for 2010. For NO_2 the yearly differences are minor.

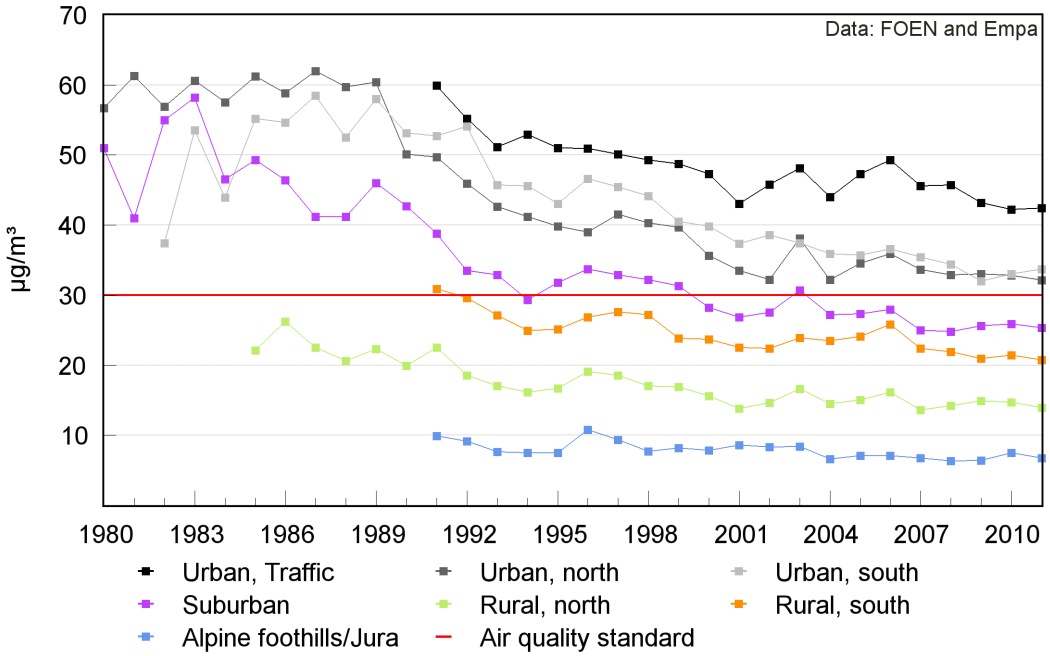

Figure 29: Annual mean NO_2 concentrations for Switzerland. From FOEN (2012).

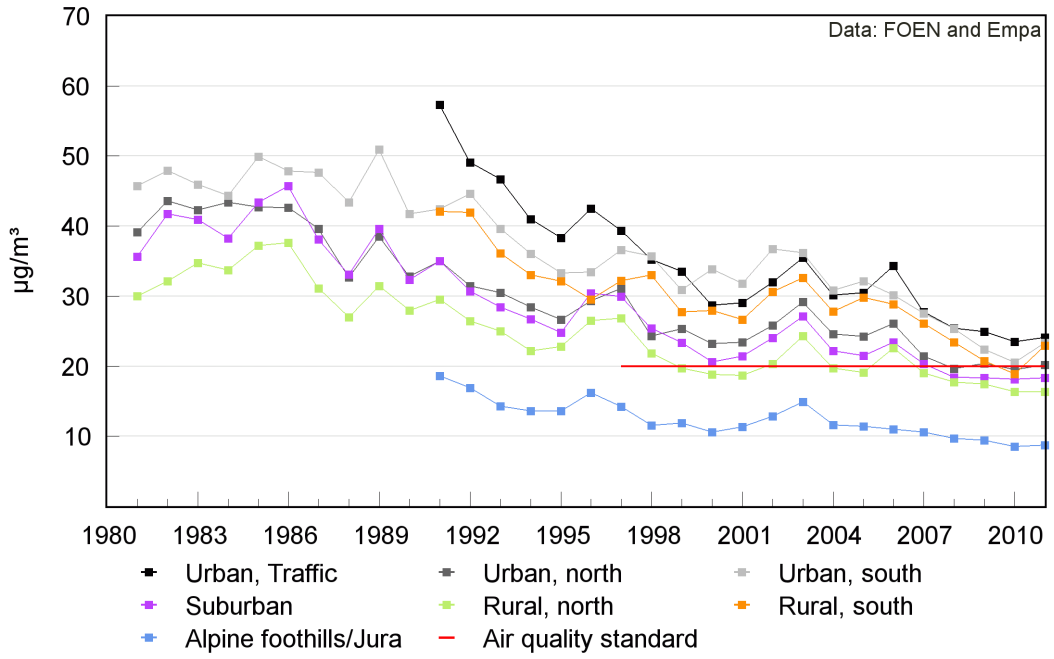

Figure 30: Annual mean PM_{10} concentrations for Switzerland (calculated from TSP prior to 1997). From FOEN (2012).

4. Summary and conclusions

This part summarizes the discussions and conclusions mentioned in the previous chapters and gives a critical view of the methods. The gain of the achieved results of WRF-Chem to air pollution exposure assessment is also discussed. After assessing the outlook for further improvements and possibilities, the chapter ends with overall conclusive remarks.

SAPALDIA is a longitudinal Swiss cohort study on air pollution and health effects which was conducted in the years 1991, 2002 and 2010. The annual averaged population based air pollution exposure are the most important statistical values for a longitudinal study. WRF-Chem achieved very satisfactory results for yearly air pollutants with Pearson correlation coefficients up to 0.8. With increased horizontal resolution, the meteorological and local geographic parameters are getting more important and good results are more difficult to achieve. Spatial patterns of O_3 and NO_2 are mainly controversial. Ozone concentrations are typically high in mountainous regions and low in the bigger cities and in the Swiss Plateau. PM_{10} is advected from highly industrial parts of northern Italy to the canton of Ticino and surroundings. All three modeled years (1991, 2002 and 2010) showed similar pattern beside local point sources which were added to the anthropogenic emissions for 2010. Maps with differences between the model and observations reveal minor discrepancies at some border regions (e.g. Geneva or Basel). However, the largest differences appear often at individual stations which is explainable with the geographical characteristics of the measurements. These measurement stations reside in narrow valleys, urban street canyons or beside heavy traffic roads and freeways. The model with its horizontal resolution of 2 by 2 km^2 cannot represent such local characteristics.

The only comparison for the specific region of Switzerland could be made with the results of a dedicated dispersion model (PolluMap). No other chemical transport model was run for Switzerland for such long periods as done in this study. WRF-Chem achieved similar results to PolluMap for PM_{10} and had only slightly decreased statistical values for NO_2. However, in addition to yearly averaged concentrations of particulate matter and NO_2, WRF-Chem is capable of producing hourly values for over 60 pollutants. Not even measurements can be used for such broad applications. O_3 concentrations could for example be simulated with R-values up to 0.75 for daily averages. The hourly temporal resolution of WRF-Chem allows the assessment of seasonal and diurnal patterns as well as weekend/weekdays differences. Diurnal cycles show the same trends for the years 2010 and 2002. Ozone concentrations were slightly higher in 2010 and NO and PM_{10} slightly lower compared to 2002. In comparison to measurements, the model has smaller amplitudes and not all peaks are captured satisfactory. This shows that the model is not capable of simulating peak values satisfactory. Special post-processing routines focusing on daily maxima would be needed, if the interests of the study were predicting violations of the air quality standards as set by the Swiss government and the European commission.

The first results showed that it is mandatory to apply a post-processing routine to achieve good results in comparison to measurements. Without any corrections, only particulate matter (PM_{10} & $PM_{2.5}$) was more or less in the correct range. Nitrogen oxide (NO_2 & NO) was under-predicted and ozone over-predicted. A simple bias offset correction already leads to a significant improvement. However, the best results were achieved with a multi-linear regression, which was applied for different categories. A lot of temporal highly resolved measurements are needed to apply such post-processing routines. The more measurements available, the more different categories can be separated which further improves the results. Most important predictors for the regression were meteorological parameters as well as seasonal functions (modified sine and cosine functions). Overall, the correlation factors for all pollutants increased due to post-processing. However, spatio-temporal differences between the year 2010 and 2002 revealed, that raw model output better matches the change over the years seen in categorized measurements. Interestingly, almost the same results could be achieved with the raw model output of the European domain combined with meteorological parameters from the Swiss domain. As the chemical part of WRF-Chem is by far the most computationally expensive, a lot of computer resources could be saved by applying a post-processing of the European chemical results with the Swiss meteorological parameters.

Nevertheless, to calculate the Swiss domain, the European domain is mandatory to get the necessary initial and boundary conditions (IC/BC's). European results showed good results for observed air pollutants, with O_3 being the most and PM_{10} being the least satisfying. As the European domain has not been post-processed, the results have some bias. NO_2 concentrations were under-predicted and daily trends of PM_{10} could not all be represented by WRF-Chem. This work briefly compared the European results of WRF-Chem to other available models already applied for Europe. We have to take into account that different models are not directly comparable. However, a comparison of Pearson correlation coefficients revealed that WRF-Chem is one of the best models for simulating air pollutants on the European scale.

It was important to test different options of IC/BC's for the European domain. Because on one hand, GFS data is simply not available for the years 1991 and 2002 and on the other hand because the standardized chemical profile of the NALROM model lacks spatial as well as a temporal resolution. It appears that chemical BC's mainly have an influence on O_3. The biggest influence was near the tropopause as the LMDZ-INCA model represents the stratosphere with higher O_3 values than NALROM. The availability of horizontal spatial differences also led to different influences on near-ground O_3. The influence of different chemical BC's on NO_2 is negligible and the influence on PM_{10} could not be evaluated as the LMDZ-INCA model does not include particulate matter. Meteorological IC/BC's had almost no influence on O_3 and NO_2 and a small influence on PM_{10}. However, Reanalysis II datasets are a good alternative for the years before 2004 when GFS simulations were absent. According to Szopa et al. (2009) better results can be achieved with even more refined chemical BC's. However, the influence is minor

for the Swiss domain as it gets its IC/BC's directly from the coarser European domain. The best results for the European domain were achieved with chemical BC's from the LMDZ-INCA model and meteorological IC/BC's from the GFS model.

Overall, WRF-Chem with the application of post-processing routines performed very good for rural, sub-urban and urban background areas. Best correlations with measurements were achieved for daily mean ozone values and yearly mean NO_2 and PM_{10} values.

4.1. Study limitations

All chemical transport models are highly depended on a good emission basis to achieve good results. The anthropogenic emissions for this study were mainly based on a top-down approach with annual values for the whole of Switzerland which were reallocated according to landuse categories. This is mainly due to the lack of more refined information on local emissions. A better solution would be the strategy of a bottom-up approach. This was already done for anthropogenic emissions from roads and by adding point sources to the 2010 emissions. However, a dedicated emission model with a bottom-up approach for almost all categories would be meaningful for further studies. Without measurements, no validation of the model could be done. The more temporally high resolved measurements available, the better the actual validation and post-processing routines. In this study, a reasonable multi-linear regression could not be done for PM_{10} and NO for the year 1991 as there simply are too few valid measurements. It is for this same reason that the main focus of this study was on pollutants where measurements are at least partially available. One of the biggest limitations of this study approach is the high demands on computational resources and programming skills. An increase of the horizontal resolution of WRF-Chem to 1 by 1 km^2 would ultimately lead to an increase of computing resources by a factor around 10. Even though the availability of computing resources increased rapidly over the last decades, the CTM's themselves have limitations and cannot use the computing resources to their full extend. This leads to an overall increase of computing time. Programming skills are needed if the source code has to be changed. For this study, this had to be done for the topography smoothing as the WRF-ARW dynamical core had stability problems over Switzerland for the horizontal resolution of 2 by 2 km^2. Minor limitations are the incompatibility of some chemical options as indirect and direct radiative feedback as well as wet scavenging with chosen physics options. Another issue is the lack of a secondary organic aerosol module for MOSAIC in the version of WRF-Chem used.

4.2. Relevance to exposure assessment

It is a novelty that a chemical transport model (CTM) is used for a longitudinal epidemiological study with three complete modeled years. Liu et al. (2007, 2012)

already gave an overview of already mentioned techniques for population based air pollution exposure. At this point in time, it was already exceptional to use a specific dispersion model for such applications. However, the simulations of WRF-Chem open new possibilities for epidemiological studies. One benefit of the very high temporal resolution is the calculations of diurnal cycles and differences of weekdays and weekends. But these two products are very sensitive to anthropogenic emissions. For the simulated years, the differences of weekdays and weekends emissions were just split up as fractions and therefore not very insightful. Overall, the availability of daily mean and monthly mean values beside annual means can lead to better personal air pollution exposure estimates. WRF-Chem is based on physically resolved meteorology and parameterized chemical reactions. Therefore the output consists of almost all possible air pollutants. Compared to PolluMap or central-site measurements, this is a big advantage. However, to really use the output of CTM's, a validation should be conducted for each pollutant. This is not possible due to the lack of such extensive measurements. The calculation of meteorological parameters are useful for post-processing routines. No assumptions of the meteorological or climatological conditions have to be made.

The usage of CTM's for epidemiological studies also has some downsides. For small studies, the skill effort and the needed computational resources do not justify the implementation of CTM's. However, once such a system is operational it can be used for simulating other years or for analyzing specific hypotheses. The limits of the spatial resolution of such models is approximately 1 km. Therefore in-city gradients (e.g. street canyons) are not captured by such models. Urban background levels on the other hand can be reproduced satisfactorily. The difference of personal exposure between home and workplace are only significant if they are separated by more than five grid points (e.g. 10 km for the results of this study). Even then differences will not be huge as the pollutants are dispersed and advected by meteorological parameters. All CTM's just simulate outdoor exposure and are not replacing indoor measurements or special indoor exposure models.

As mentioned before, WRF-Chem is good for reproducing background levels of all different categories for Switzerland. Additionally the model results can be used as input data for more refined models. Chap. 3.3 showed the differences of the raw model output and the post-processed results. If the simulated years are thought for individual air pollution exposure, the post-processed data should be used. They better represent the measurements compared to the raw model output and have overall better absolute values. However, if the model results are further post-processed (e.g. as input for other models) or if only the spatio-temporal differences are of interest, the raw model output is the better choice. How to use these kind of CTM's results are elaborated in the following section.

4.3. Outlook

WRF-Chem is frequently updated and still under development. This allows the implementation of additional sub-modules or further studies of specialized topics. For example, ultrafine particles are getting more and more important in regards of health effects (Seaton et al., 1995; Peters et al., 1997; Ibald-Mulli et al., 2002; Nel, 2005). An output of particulate matter smaller than 1 μm could be implemented in the output through a modification of the aerosol module. A newly developed secondary organic aerosol (SOA) module has been added to WRF-Chem from version 3.3 onwards (Grell et al., 2011*b*). Hüglin et al. (2005), Volkamer et al. (2006) and Hallquist et al. (2009) show the increasing importance of the formation of SOA to the concentrations of $PM_{2.5}$ and also for human health effects. Therefore, a new short evaluation study including SOA should be done. As mentioned before, a dedicated Swiss emission model as well as more refined chemical BC's for the European domain would also benefit the overall results. Overall, WRF-Chem is good for producing background values for the whole of Switzerland. However, the model is not capable of predicting within-city differences due to the limitation of the horizontal resolution. The gridded output of WRF-Chem can serve as input for other more refined models like landuse regression (LUR) or computational fluid dynamics (CFD) models. Another possibility would be to combine data from satellites or other remote sensing applications (e.g. Popp et al. (2012)) with the results of WRF-Chem. A similar approach with a LUR model and satellite data was done by Novotny et al. (2011). The Environmental Software and Modelling Group of the Technical University of Madrid has even implemented WRF-Chem in their air pollution real time forecasting system (San José et al., 2008; Kukkonen et al., 2012).

All the above mentioned applications of WRF-Chem suppose a high-resolution domain. This leads ultimately to high demands on the model expertise and on the computational resources. However, an easier approach was briefly explained. If the post-processing routines are trained with high resolution meteorological parameters and coarser chemical results, almost the same results are achieved. This approach is worth further investigation. There are already many operational meteorological forecast models with a high horizontal resolution. Without any additional computing resources used, these outputs could be combined with coarser chemical runs or even already existing global chemical models.

4.4. Conclusive remarks

This study showed a new approach to get population based air pollution exposure. This paper has developed such a modeling system, evaluated the results, compared the system to other models, provided a critical view and evaluated the relevance of this study to health assessments. The application of WRF-Chem needs a high skill effort and high computing resources and is strongly dependant on good anthropogenic emissions.

Overall, WRF-Chem performed satisfactorily for background levels. Compared to approaches in Liu et al. (2007) many advantages could be listed. The possibility to use the results as input for other more refined models (LUR, CFD) is especially important. As an outlook, an application of WRF-Chem with special post-processing routines between high resolution meteorological parameters and coarser chemical models is mentioned. To conclude, WRF-Chem is very useful for larger epidemiological studies or for specialized sub-topics.

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Appendix A Chemical conversion factors (EMEP to CBM4)

Pollutant	SNAP 1	SNAP 2	SNAP 3	SNAP 4	SNAP 5	SNAP 6	SNAP 7	SNAP 8	SNAP 9	SNAP 10	SNAP 11	mol. weight
CO	1.0000	1.0000	1.0000	1.0000	1.0000	0.0000	1.0000	1.0000	1.0000	1.0000	0.0000	28 g/mol
NO	0.6200	0.6200	0.6200	0.5869	0.5869	0.0000	0.6200	0.6200	0.6200	0.5869	0.0000	30 g/mol
NO2	0.0500	0.0500	0.0500	0.1000	0.1000	0.0000	0.0500	0.0500	0.0500	0.1000	0.0000	46 g/mol
SO2	1.0000	1.0000	1.0000	1.0000	1.0000	0.0000	1.0000	1.0000	1.0000	1.0000	0.0000	64 g/mol
NH3	1.0000	1.0000	1.0000	1.0000	1.0000	0.0000	1.0000	1.0000	1.0000	1.0000	0.0000	17 g/mol
FORM	0.0083	0.0083	0.0083	0.0083	0.0005	0.0000	0.0008	0.0057	0.0000	0.0005	0.0000	-
PAR	0.0239	0.0239	0.0239	0.0270	0.0364	0.0277	0.0279	0.0146	0.0011	0.0270	0.0000	-
TOL	0.0001	0.0001	0.0001	0.0006	0.0000	0.0007	0.0013	0.0001	0.0000	0.0006	0.0000	-
ALD2	0.0000	0.0000	0.0000	0.0008	0.0000	0.0017	0.0008	0.0023	0.0000	0.0008	0.0000	-
ETH	0.0000	0.0000	0.0000	0.0013	0.0000	0.0006	0.0019	0.0062	0.0000	0.0013	0.0000	-
OLE	0.0000	0.0000	0.0000	0.0019	0.0000	0.0026	0.0011	0.0029	0.0000	0.0019	0.0000	-
XYL	0.0000	0.0000	0.0000	0.0004	0.0000	0.0006	0.0010	0.0001	0.0000	0.0004	0.0000	-
ISOP	0.0000	0.0000	0.0000	0.0001	0.0000	0.0002	0.0000	0.0000	0.0000	0.0001	0.0000	-
PEC	0.2760	0.0934	0.0000	0.0183	0.0000	0.1000	0.1901	0.6587	0.0350	0.0750	0.0000	-
PMFINE	0.1497	0.3386	0.2751	0.8367	0.7202	0.3995	0.2079	0.0000	0.8902	0.2644	0.0000	-
PNO3	0.0025	0.0024	0.0055	0.0035	0.0028	0.0005	0.0014	0.0032	0.0000	0.0063	0.0000	-
POA	0.3106	0.3155	0.4200	0.1102	0.0420	0.4800	0.5682	0.2921	0.0068	0.6389	0.0000	-
PSO4	0.2611	0.2501	0.2995	0.0313	0.2350	0.0200	0.0324	0.0460	0.0680	0.0154	0.0000	-
PM10	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	0.0000	-
RtTe	1.7350	1.7350	1.7350	1.0000	1.1200	1.0300	1.0000	1.0000	83.3300	1.0000	0.0000	

All calculations are done for each SNAP sector and per pollutant. The input into WRF-Chem for pollutants is in $^{mol}/_{h \cdot km^2}$ and in $^{\mu g}/_{s \cdot m^2}$ for PM.

CO, NO, NO_2, SO_2, NH_3:

$$E[^{mol}/_{h \cdot km^2}] = \frac{E[^{Mg}/_{h}] \cdot fac \cdot 10^6}{mol.weight \cdot (10^{-3} \cdot dx)^2}$$

FORM, PAR, TOL, ALD2,
ETH, OLE, XYL, ISO:

$$E[^{mol}/_{h \cdot km^2}] = \frac{E[^{Mg}/_{h}] \cdot fac \cdot 10^6 \cdot RtTe}{(10^{-3} \cdot dx)^2}$$

PEC, PMFINE, PNO3, POA,
PSO4, PM10:

$$E[^{\mu g}/_{s \cdot m^2}] = \frac{E[^{Mg}/_{h}] \cdot fac \cdot 10^{12}}{3600 \cdot dx^2}$$

E: Emissions
fac: factors from the table
RtTe: additional conversion factors for lumped chemicals
mol. weight: molecular weight
dx^2: grid spacing of the emissions in meter

Conversion factors developed by the Earth Science Group of the Barcelona Supercomputer Center (EPA, 2010; Parra, 2004; Arévalo, 2005).